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# RADIONUCLIDE ANALYSIS OF ENVIRONMENTAL SAMPLES. A LABORATORY MANUAL OF METHODOLOGY

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Cincinnati, Ohio

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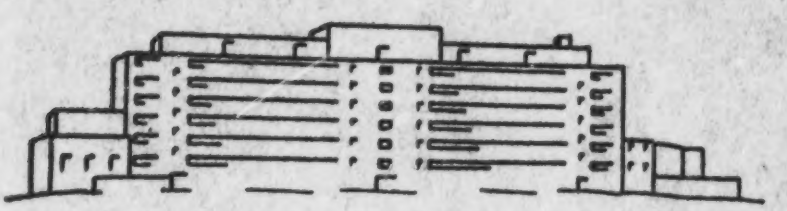

# RADIONUCLIDE ANALYSIS OF ENVIRONMENTAL SAMPLES
## A Laboratory Manual of Methodology

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

# RADIONUCLIDE ANALYSIS
# OF
# ENVIRONMENTAL SAMPLES

Paul F. Hallbach, Editor

November 16, 1959

U. S. Department of Health, Education, and Welfare

Public Health Service

Bureau of State Services

Division of Radiological Health

Robert A. Taft Sanitary Engineering Center

Cincinnati 26, Ohio

## FOREWORD

This manual summarizes the analytical methods that have been developed by the staff of the Robert A. Taft Sanitary Engineering Center or modified from methods developed by others for use with environmental samples. The methods given provide procedures for identification of many radionuclides in a wide variety of environmental media.

Brief statements included on instrumentation are of a general nature only and will soon be supplemented by a more extensive compilation of our instrumental techniques.

As new methods are developed or existing methods revised, these will be made available to the recipients of the previous releases so that they may keep their manual current. For this reason the loose-leaf format has been adopted for the manual.

Any suggestions or comments regarding the methods included in the manual or difficulties encountered as a result of applying the techniques to other types of samples should be brought to the attention of our staff at Cincinnati.

Conrad P. Straub

Conrad P. Straub, Chief
Radiological Health Research Activities
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# I. INTRODUCTION

## INTRODUCTION

In this introduction an attempt is made to describe some of the general principles underlying radioactivity measurements, as well as to give some comments on the general philosophy used in the Radiological Health Research Activities Program of the Division of Radiological Health at the Robert A. Taft Sanitary Engineering Center.

## GENERAL PRINCIPLES OF RADIOACTIVITY COUNTING

The sensitivity and precision of radioactivity counting measurements is limited by the statistically random nature of radioactivity. Radioactive disintegrations occur randomly so that the counting rate of any sample fluctuates about its mean.

Because every radiation measuring instrument has a background counting rate even in the absence of any sample, it is necessary to determine a counting rate as a difference between an observed (sample plus background) counting rate and the background counting rate. From this it follows that the standard deviation s of the net counting rate is described by this equation:

$$s = \left( \frac{R_o}{t_o} + \frac{R_b}{t_b} \right)^{1/2} \qquad 1)$$

In this equation the R's are the counting rates, the t's are the times of observation, the subscript "o" refers to the observed or sample plus background measurement and the subscript "b" refers to the background measurement. This equation may be written in terms of R, the net counting rate and $R_b$, the background counting rate through the following steps:

$$s = \left( \frac{R + R_b}{t_o} + \frac{R_b}{t_b} \right)^{1/2} \qquad 2)$$

$$s = \left( \frac{R + R_b}{t_o} + \frac{(t_o/t_b)\, R_b}{t_o} \right)^{1/2} \qquad 3)$$

$$s = \left( \frac{R + \left(1 + \frac{t_o}{t_b}\right) R_b}{t_o} \right)^{1/2} \qquad 4)$$

The fractional counting error, $\frac{s}{R}$, is then given by the following equation:

$$\frac{s}{R} = \left[ \frac{1 + \left(1 + \frac{t_o}{t_b}\right)\frac{R_b}{R}}{R\, t_o} \right]^{1/2} \qquad 5)$$

Although this is a rather formidable expression, there are two simplifying cases. The first of these, when $R_b << R$, reduces to

$$\frac{s}{R} = \left( \frac{1}{R\, t_o} \right)^{1/2} \qquad 6)$$

In this high activity case the fractional error is independent of background. The other simplification occurs when $R_b >> R$. In this case the equation reduces to

$$\frac{s}{R} = \left[ \frac{\left(1 + \frac{t_o}{t_b}\right)\frac{R_b}{R}}{R\, t_o} \right]^{1/2} \qquad 7)$$

This can be written as

$$\frac{s}{R} = k \left( \frac{R_b}{R^2} \right)^{1/2} \qquad 8)$$

In this extremely low activity case the error is dependent on the first power of the net counting rate R and on the square root of the background counting rate.

We thus see that even in low activity measurements the net counting rate is more important than the background counting rate. In order to obtain a large counting rate two methods are used. One is to use a large sample. The second is to detect the radiation emitted from this sample with high efficiency. Reduction of background, while important, is less significant than either of these first two factors.

In accordance with these principles, the methods given are normally applicable to samples at least as large as a liter or kilogram and high efficiency instrumentation is used throughout. Thus, the most common instruments used for alpha and beta measurements are the internal proportional counter which may have efficiencies ranging from 50 per

cent for alphas to as high as 75 per cent for hard betas. The relatively high beta backgrounds of these instruments (around 50 to 60 cpm unshielded and 20 to 25 cpm shielded) do not become a limiting factor until very low counting levels are reached. For extremely low counting rates the low background (anti-coincidence) beta counter is preferred. Although this instrument has a lower efficiency than the internal proportional counter (42 per cent for yttrium-90 as compared with about 75 per cent for the internal counter) the extremely low background (about 0.6 cpm) more than makes up for this. This can be more specifically stated in terms of the figure of merit which can be defined as the efficiency over the square root of the background for very low level counting. The figure of merit for the internal counter is

$$\frac{0.75}{60^{1/2}} = 0.097$$

for the unshielded counters, approximately

$$\frac{0.75}{25^{1/2}} = 0.15$$

for the shielded internal counters and about

$$\frac{0.42}{0.6^{1/2}} = 0.54$$

for the low background beta counter. The other commonly used instrument is the low level gamma spectrometer which is used with as large samples as are available and has a large sodium iodide crystal for high efficiency together with a heavy shield for minimum background.

## GENERAL PHILOSOPHY OF RADIONUCLIDE ANALYSIS

The general philosophy in analytical radionuclide methods developed by Radiological Health Research Activities has been to make the maximum use of both chemistry and physics so as to minimize sample preparation and to obtain the most information from a given sample. Maximum advantage is taken of modern methods of counting, such as gamma spectroscopy.

The low level gamma spectrometer, as mentioned before, is the backbone of the instrumental techniques for radionuclide analysis. This instrument, although expensive, earns its keep in several ways. First of all, no sample preparation is required. This not only results in a saving of manpower but avoids the possibility of introducing error through losses in manipulation or accidental contamination. Another virtue of the gamma spectrometer is that it permits the determination of several

radionuclides in a single count. Thus, in milk samples, the three gamma-emitting isotopes, iodine-131, cesium-137, and barium-140 - lanthanum-140 are determined in a single 100-minute count. Gamma spectroscopy is also used as a guide in radionuclide separations. A preliminary gamma spectrum determination may disclose the presence of unsuspected radionuclides. In environmental samples these have included the non-fission products, zinc-65, cobalt-60, cesium-134, and tungsten-185.

Not only is gamma spectroscopy used as a guide in chemical separations but conversely chemical group separations are often used as a means of obtaining gamma spectra sufficiently simple for interpretation. In this work the conventional qualitative analytical schemes are usually employed, with such steps as precipitation of sulfide from acid solution, precipitation of sulfide from basic solution, precipitation of oxalates, and the like being important.

Unfortunately, not everything can be done by gamma spectroscopy. A number of the more important radionuclides, such as strontium-89 and strontium-90, have no gamma radiation. When a substance is a gamma emitter radionuclide separation may be necessary, either for greater sensitivity or to obtain a simpler spectrum. Thus, for example, radium-226, although in principle it could be determined through the gamma emissions of its daughters, is usually measured by alpha counting where the combination of high efficiency and low background make for maximum sensitivity.

Much of the early work on radiochemical separations, as exemplified by the collection of methods edited by Coryell and Sugarman, was developed for relatively high level solutions where thousands to millions of disintegrations per minute per ml were available. Furthermore, in many of these determinations the detection of short-lived radionuclides was of considerable importance. Consequently, it is not strange that the methods developed used relatively small samples, rather rapid low-yield methods, and conventional instrumentation. In measuring environmental samples we have had to depart from this philosophy. Because of the low levels sought in many of our samples, we have been compelled to take large samples and to process them in such a manner as to obtain the radionuclide interest in high yield. Since the samples in most cases are sufficiently old upon receipt that they do not contain significant quantities of short-lived radionuclides, relatively slower but more quantitative methods have been used. The large samples -- of the order of liters or kilograms of material -- necessarily introduce large amounts of nonradioactive contaminants such as calcium, silica, and organic matter. The methods have had to take these inert contaminents into account. Wherever practical, we have not made a quantitative separation of these stable components. This has made it impossible to determine chemical yields by weighing and we have preferred instrumental techniques, such as flame or absorption spectrophotometry, or else have depended upon a high relatively reproducible yield without actually determining it for each sample.

Both the purification procedures and the final sample preparation techniques have been simplified wherever possible so as to take advantage of modern instrumentation. For example, in the determination of cesium, if the final counting is with a gamma spectrometer, the separation of nuclides which do not interfere with the gamma count (either through absence of gamma radiation or because their gamma energy is quite low), is not essential. Thus, relatively large quantities of the strontium isotopes or the cerium isotopes can be tolerated in the final cesium fraction. On the other hand, good decontamination from such materials as zirconium-95 - niobium-95, whose 0.72 to 0.76 mev gamma radiation interferes strongly with the 0.66 mev radiation of the cesium, is essential.

SAMPLING

Analytical results are only as good as the samples upon which the analyses are made. If the samples are representative, the results will be; if not, the analyses will be non-representative. The sample must also be adequate for the kind of analysis required. Grab samples are useful for measuring fluctuations in activity levels and to identify short-lived radionuclides. Composite samples represent average conditions covering the period of sampling, whereas continuous samples provide information of a dynamic nature and allow evaluation of environmental effects over a period of time.

The size of sample required is determined by the levels of radioactivity and the number and kind of radiochemical separations to be performed.

A.S.G.
11-16-59

# II. INSTRUMENTATION

## GENERAL PRINCIPLES

There are basically three types of radiation which we are principally interested in measuring. They are: 1) alpha, 2) beta, and 3) gamma radiations. Each requires a different type detector for maximum counting efficiences.

The alpha particles are comparatively large and heavy, high-energy particles which do not have much penetrating power. It is therefore necessary to have a minimum of absorbing material between the source and the sensitive part of the detector. Internal counters are therefore ideal for alpha detection. An activated zinc sulfide phosphor is also a very good detector for this form of radiation. It is important that the solid content of the samples be kept close to negligible to reduce self-absorption losses.

Although absorption losses are not as serious for beta counting as in alpha detection, they nevertheless must be taken into account and corrections made. It is necessary to know the maximum energy of the beta, the thickness of the sample, and the type of backing the sample has. All these factors enter into the determination of the disintegration rates of betas. The energy of the emitter determines the type of detector to be used. If the betas are of very low energy, such as carbon-14 or tritium, the absorption losses in most samples are severe, and specialized instrumentation is necessary to obtain accurate counting.

Gamma absorption is very small and consequently self-absorption losses in the sample are not serious. Since this is true, large samples may be employed. The gamma interaction with a counter is proportional to its mass. For spectroscopy, large detectors of high atomic weight are most efficient. Sodium iodide crystals, thallium-activated, are commonly used for this purpose.

Counters which use windows as their means of letting radiation into the sensitive part of the detector have some inherent limitations. Alpha and low-energy beta radiations are partially lost, particularly if the windows are over 1 mg/cm$^2$ or the path distance is greater than one-half inch. Since the geometry factor is very important in connection with counting efficiency, it is pertinent to place the source as close to the detector as possible unless an extremely radioactive sample is to be determined.

Proportional counters have the advantage of being able to discriminate between alpha and beta-gamma radiations. By simply lowering the operating voltage, only alphas will be counted and with higher voltages, alphas plus betas and gammas are detected. The alpha particles create more ion-pairs in passing through the gas and consequently produce

larger pulses. It is therefore possible to count only alpha by lowering the operating voltage to the plateau for these heavy particles. At the lower operating voltage the proportional counters are insensitive to beta and gamma radiations.

Scintillation counters are finding more and more acceptance since satisfactory photomultiplier tubes have been developed. Depending upon the type of detector, they can be used for alpha, beta, or gamma radiation. A zinc sulfide screen, insensitive to beta or gamma rays, is used for the detection of alphas. Beta scintillation counters commonly use anthracene or a plastic scintillator as the detector, and they can be made relatively insensitive to gamma rays. A sodium iodide crystal is used for detecting gamma rays since it is made of material having relatively high density. Alphas and betas are stopped by a can which is required in any event to exclude atmospheric moisture.

## LOW LEVEL WORK

When measurements need to be made on samples having little activity, the instrumentation becomes more complex. First of all, it is essential to count as many of the radiations given off as possible, since there are very few in the sample. To accomplish this, the geometry of the sample to the detector must be good to obtain as high an efficiency value as possible. Windows should be absent or very thin. In addition to this, it is important to choose as large a sample as possible. Furthermore, the background radiation should be minimized. This is accomplished by heavy shielding of the detector with high density material and/or electronic shielding using coincidence techniques.

## NUCLIDE IDENTIFICATION

Instrumental identification of the specific nuclides present in environmental samples is becoming increasingly important. As an initial approximation, alphas can be distinguished from beta-gamma radiations. Isotopic analysis can be done by spectroscopy, or by the determination of the maximum beta energies. In both cases, by knowing the energies of the radiations, the nuclides can be determined through their decay schemes.

G.J.K.
11-16-59

# SPECIFIC INSTRUMENTS

## SPECIFIC INSTRUMENTS AND THEIR APPLICATIONS

All the instruments described in this section have been shown and elaborated on in an instrumentation manual, Robert A. Taft Sanitary Engineering Center Report R59-3.

## GEIGER-MUELLER COUNTER

The conventional G-M counter tube has largely been superseded by better instrumentation, especially for the measurement of typical environmental samples. The low detection efficiency and the unsuitability for large samples limit its use. In a 1-1/2" thick lead shield a typical tube will have a background of about 30 cpm. Since the sealing window must be about 2 mg/cm$^2$, there is a substantial absorption loss for alphas and betas.

To insure that the G-M detector is functioning properly, it be plateaued. The plateau should not have a slope of more than 2 per cent per 100 volts and it should be about 250 volts long for a new tube. As the tube ages, the plateau will shift and probably shorten. Once the operating voltage has been determined, the tube should be checked by counting a source each morning, noon, and night to insure its functioning properly. Background measurements should also be made at least twice a day.

## INTERNAL PROPORTIONAL COUNTER

The internal proportional counters are a useful tool for measuring levels of activity commonly found in environmental samples. They are called internal because the sample is placed into the sensitive part of the detector and counting gas purged throughout the counter and sample. Since the source is within the counter, the radiations only have to penetrate the sample itself and produce several ion-pairs in the gas for detection. The locations of the activity in the planchet is not a serious problem since the counter is almost equally sensitive over the 2" diameter.

The counters are called proportional because the size of the pulse produced is proportional to the number of ion-pairs produced. Since alphas dissipate much energy in the chamber, they produce more ion-pairs than betas and consequently make larger pulses. By operating at a relatively low voltage, which gives a small amplification, the large alpha pulses can be detected while the smaller beta-gamma pulses are discriminated against. Ability to count alphas only at one voltage, and alphas plus beta-gamma at another, makes the proportional counter a very versatile detector.

For an unshielded 2-1/4" diameter hemisphere and piston, the beta background is about 50 - 60 cpm and for a shielded chamber (2" of lead), it is about 30 cpm. Alpha backgrounds are about 0.2 cpm. Background counts should be taken at least twice a day to check for possible contamination.

The instruments should be plateaued with extended sources, 2" in diameter, with plated uranium oxide ($U_3O_8$) or equivalent. A source of this type most closely simulates samples and should be used in preference to a point source. This emitter can then be counted several times throughout the day to check out the instrument on a day-to-day basis. An uncovered uranium oxide source will serve as the alpha source and an aluminum covered one for the beta. The counting efficiency for betas is about 55-75 per cent with negligible solids, depending on energy. The alpha efficiency is approximately 50 per cent for weightless samples.

## END WINDOW PROPORTIONAL COUNTER

For 7" x 9" filters, used for air sampling, a 7-1/2" diameter proportional counter with a very thin Mylar window is used. Due to the large area of the detector, it must be purged with counting gas for approximately one hour before it can be used. The instrument is then checked by a standard to see if the counting rate is sufficient and if it is, a background count is made and the counter is ready for use.

Since the counter is so large, the background for an instrument of this type, without shielding, is around 500 cpm and the plateau is about 150 volts. It is very useful because it enables large samples to be counted and it reduces the self-absorption loss in the sample because it can be spread out thin.

Proportional counting gas is used because it enables high activity rates to be counted without excessive dead time losses.

## LOW BACKGROUND (ANTI-COINCIDENCE) BETA COUNTER

A counter consisting of central detectors with 2.5 tons of steel shielding and an umbrella of Geiger tubes comprises a low level beta counter. The steel stops most of the external gammas and the umbrella is in coincidence with the central counters used for the samples. This electronic shielding blanks out any radiations which interact in both the umbrella and the central counter, thereby eliminating cosmic ray interference. The combination reduces the background for the 2" end window tubes to less than 1 cpm.

The 0.9 $mg/cm^2$ Mylar window is so thin a continuous gas purge is necessary. This end window counter has the advantage of keeping

contamination at a minimum but is still thin enough to prevent serious attenuation of betas above 500 Kev. With the extended 2" diameter source close to the window the counting efficiency for yttrium-90 on stainless steel is 42 per cent. Lower energy beta emitters count with lower efficiencies.

## LIQUID SCINTILLATION COUNTER

This instrument is particularly well suited for counting low energy beta emitters such as tritium or carbon-14. By placing the sample in solution in a liquid scintillating medium, self-absorption losses are eliminated and the counting efficiency can approach 100 per cent. It is not limited to low energy beta work, but it, or a method equivalent to it, is essential for these weak emitters. Sample preparation is the big drawback for a counting technique of this type.

By placing two photomultiplier tubes in coincidence and cooling them in a deep freeze, the background, due to dark current in each tube, is nearly eliminated. Counting efficiencies are dependent on the type of solvents used and also the energy of the beta.

Since the detector is connected to a two-channel analyzer, specific nuclides can be determined simultaneously by counting in certain energy regions. It is therefore possible to count both tritium and carbon-14 in the sample at the same time.

## ALPHA SCINTILLATION COUNTER

The alpha scintillation detector is composed of a zinc sulfide screen and photomultiplier tube housed in a light tight box and coupled through a cathode follower into a scaler. The alpha source is placed as close to the screen as possible without undue risk of contamination to prevent absorption losses in the air path. Close to the screen the counting efficiency is over 40 per cent.

This counter is useful because it does not require a gas supply nor does it matter if the sample is not firmly held in place as would be the case in the internal counters. It is handy and easily set up for field use.

The background is about 0.2 cpm for a 3.5 inch screen, and the plateau is about 200 volts, but it is dependent on the photo-tube.

## GROSS GAMMA COUNTER

A 2" sodium iodide crystal and photomultiplier assembly fed through a cathode follower is coupled directly to a scaler for gross gamma measurements. Either a solid or well crystal can be used for this purpose depending on the sample size.

About 1-1/2 - 2 inches of lead shielding is essential to reduce the background gamma radiation. With this amount of shielding the background is approximately 300 cpm for a 2" solid sodium iodide crystal. This gross gamma counter is very useful for tracer levels and where it is not necessary to determine the specific nuclides present in the sample. This detector may have still great applications as a means of screening out samples below a certain level.

## GAMMA SPECTROSCOPY

Extensive use is being made of gamma spectroscopy for the identification of radionuclides. With suitable calibration data, including knowledge of the effect of spectral interference, the spectrometer becomes a quantitative tool. Sample preparation is not necessary since self-absorption losses are neglible and consequently large volume samples can be scanned without concentration.

The spectrometers can be of the single or multi-channel type. These instruments sort out pulses according to energy and the multi-channel analyzer records many different energy groups at one time, whereas the single channel records only one. For samples having little activity, the multi-channel is almost essential because of the excessive counting times required to cover a spectrum with a single channel. For tracer levels the single channel is quite adequate.

The gamma spectrometers most commonly use a sodium iodide crystal optically coupled to a photomultiplier tube. The latter is connected through a cathode follower and linear amplifier into the analyzer, which records each pulse into its proper channel. Each channel represents a certain amount of energy absorbed in the crystal. The spectrum shows characteristic peaks at the full energy of the gamma ray. By knowing the decay schemes of radionuclides, it is therefore possible to identify specific isotopes by the characteristic gamma peaks.

The background and efficiency are dependent on the size of the crystal. The smaller the crystal the less the background will be for the equivalent amount of shielding, but the counting efficiency will be less. The detector is energy dependent; the efficiency being lower for higher energies. This is especially notable in pulse height analysis because it is necessary for the entire photon energy to be spent in the

crystal to be recorded under the photopeak. This type of interaction is called photo-electric and it is the most important interaction in gamma spectroscopy.

All higher energy photons interfere with lower energy spectra because of the Compton interactions in which only a portion of the total energy is absorbed in the crystal. This interference makes quantitative analysis difficult.

The resolution of the crystal and phototube combination determines how close gammas can be in terms of energy and still be identified. Resolution can be easily obtained better than 9 per cent for solid crystals and less than 10 per cent for well crystals.

G.J.K.
11-16-59

# CALIBRATION

Calibration of the instruments presents, perhaps, the biggest problem in setting up any counting tool. Each counter is energy dependent in one way or another.

It is best to obtain a calibrated standard, if available, from the National Bureau of Standards or a commercial source. Some nuclides cannot be purchased in this way, making it necessary for each laboratory to calibrate them. A four-pi beta counter has been found useful for standardization work. Sample preparation for this instrument is very exacting.

Several factors must be taken into account in calibrating alpha and beta detectors. The backing material of the sample planchet is important because the backscatter factor is a function of the atomic number of the material. The thickness of the sample is of importance because of self-absorption. Any airpath or windows between the source and the sensitive part of the counter influence the efficiency because some of the radiation is absorbed before it can be counted. The energy of the nuclide is the primary consideration, however, since all other effects depend on it.

Efficiency of detection is a function of crystal size. The number of gamma photons interacting with the crystal is proportional to crystal mass, and a greater proportion of those interacting will give up their full energy in a large crystal. This is particularly important for high gamma energies and for samples of very low activity.

It is necessary, in any event, that the calibration samples should closely resemble the actual samples as far as geometry is concerned. Since the size and geometry of the sample determines the efficiency, it is important to establish definite volumes and configurations when calibrations are made. Even the spectra itself is changed, to a certain extent, by using different sized samples.

In calibrating gamma detectors, it is necessary to know the decay scheme of the isotope to which it is to be calibrated. As an example, for each 100 dpm there may only be 10 gammas in the decay scheme, and they may have different energies. It is important, therefore, to know the abundance of each gamma to obtain quantitative results. The instrument can be calibrated in terms of counts per minute per microcurie or counts per minute per gamma photon of a given energy. If the calibration pertains to the gamma photons, a curve of efficiency vs. energy can be drawn and interpolations made between the experimental points.

G.J.K.
11-16-59

# III. GROSS RADIOACTIVITY

# ATMOSPHERIC PARTICULATE RADIOACTIVITY

## Principle of Method

Air particulates in samples are deposited on glass fiber filters by a high volume air pump. After a waiting period of at least two days after collection, the gross beta activity is measured. The samples are usually recounted after four to six days. From these two measurements, the gross beta activity on the date of sample collection and the decay rate (expressed as the half-life beginning from the date of the first count) are determinted.

## Equipment

a. Instrumentation

(1) Description: The instrument consists of a thin-window, large area, proportional gas flow counting chamber, Nuclear Measurements Corporation (Indianapolis, Indiana) Model PCC-12 having a thin (0.9 mg/cm$^2$) metal coated plastic window (Mylar). The chamber-preamplifier unit is connected to a conventional scaler (Nuclear-Chicago Model No. 181A Decade) and timer (Nuclear-Chicago Dual timer Model T1). The piston of the counter chamber is replaced with a steel plate covered with aluminum foil for ease of decontamination. The resultant shelf on which the samples are placed is 5 mm below the thin-window.

(2) Operation: Each morning the counting chamber is pre-flushed at high rate with P-10 gas (90 per cent argon and 10 per cent methane) for about an hour's time. Proper flushing is ascertained by counting the background and a secondary standard to determine whether these have changed from the previous day. Low background or standard counts indicate insufficient flushing and high background indicates contamination. Following this preliminary check of counter performance, samples are counted until noon when again the instrument is checked by counting both standard and background. This procedure is repeated at the end of the day or when all the day's samples have been processed.

(3) Calibration: The dust of a sample is deposited in an area 7" x 9" or 63 sq. in. When placed under a window having a diameter of 7.5" or 44.19 sq. in., only a portion of the

sample area is observed with a varying efficiency depending on counting chamber geometry. In order to evaluate the effective area of the counter, a point source of activity is counted in various positions from the center outward to beyond the diameter of the window. From these data the "effective area" of counting has been calculated, and determined to be 66 per cent of the sample area for the above dimensions.

By counting a source in an internal proportional counter of known efficiency, and again under the thin-window counter, it has been found that the ratio of the thin-window count to the internal proportional count is 0.86. Since the efficiency for the internal proportional counter was known, the efficiency of the air counter could be calculated. The self-absorption factor varies from 1.0 for zero thickness of sample solids to 1.2 for 600 milligrams of dust, and to 1.4 for 1000 milligrams of dust. For routine tests of the activity it is customary to assume a self-absorption factor of 1.1 for samples containing less than 601 milligrams of dust and a factor of 1.3 for samples containing from 601 to 1000 milligrams of dust.

(4) Instrument Performance: A sample of uranium oxide, plated on a 2 inch diameter stainless steel and covered with 7 milligrams per square centimeter of aluminum foil to shield out alpha activity, is used as a secondary standard for determining the plateau and checking instrument performance. These checks are made three times a day to be assured that the instrument is performing properly. The secondary standard is counted long enough to give a total of at least 10,000 counts (2 per cent precision at two standard deviations).

b. Standards Used

(1) Standards consist of extended sources (2 inches in diameter) of uranium oxide plated on stainless steel and covered with 7 $mg/cm^2$ of aluminum foil and standard solutions of thallium-204, obtained from the National Bureau of Standards and deposited on dishes. These standards are used to determine the counting efficiency of internal proportional counters and the thin-window counter.

## Procedure and Calculation

(1) The samples are counted for two 2-minute periods usually upon receipt and again after 4 to 7 days. If the rate of decay is relatively low, indicating fission products formed at least 10 days or more before the first count, the data are plotted on semi-log paper and extrapolated back to the time of sample collection. If a rapid rate of decay is observed, the results are plotted on log-log paper in such a fashion that the Hunter and Ballou decay factor of -1.2 or somewhat less is obtained and the results extrapolated to time of collection.

(2) The counting rate (R) is corrected to disintegration rate (A) by the use of the following formula:

$$A = \frac{(R)(S)}{E}$$

where:

A = activity, dpm
E = efficiency for zero weight samples
R = net counting rate, cpm
S = self absorption factor

The value (A) is converted to μμc per cubic meter, by dividing by (2.22)(sample volume, in cubic meters).

## Estimate of Data Reliability

This is somewhat variable depending on many factors, such as the accuracy of the volume of air filtered, the filter characteristics, the age of the bomb debris when first counted, the number of days from collection to date of first count and the counting error which is a function of the counting rate. For fresh bomb debris of less than 5 days of age and relatively high counting rate, it is possible that the extrapolated value could be off by a factor of two, because fractional parts of a day are not considered. For activity having an age of ten or more days the determination should be within 10 per cent of the correct value providing reliance can be placed on the volume of air sampled and uniform characteristics of the filter. With the usual background of about 500 cpm, a sample counting above 1000 cpm per sample (net) would have a raw counting error at the 95 per cent confidence level of:

$$1.96 \left(\frac{1500 + 500}{4}\right)^{1/2} = 44 \text{ cpm}$$

or 4.4 per cent, whereas the error of a sample counting 500 cpm net (equal to the background) would be:

$$1.96\left(\frac{1000 + 500}{4}\right)^{1/2} = 38 \text{ cpm}$$

or 7.6 per cent.

L.R.S.
11-16-59

# RADIOACTIVITY IN WATER, SILT, AND BIOLOGICAL MATERIALS

## Principle of Method

Water samples are filtered, and separated into suspended and dissolved fraction. The dissolved fraction is concentrated by evaporation and transferred to a planchet, dried, flamed, and counted. Silt and biological samples are dried, transferred, and fixed (when necessary) with lucite in acetone, dried, and counted.

## Procedure

a. Water Samples

(1) Filter a sample of suitable size (at least 250 ml) through a membrane filter with vacuum.

(2) Wash the filter and solids with distilled water and continue to pass air through to dry.

(3) Retain the filtrate and washings for the determination of dissolved radioactivity.

(4) Transfer filter and solids to a weighed counting dish, saturate with ethyl alcohol, and burn off the alcohol. If any of the filter remains, add a second portion of ethyl alcohol and burn it off.

(5) Flame the dish for a few seconds, then cool, weigh, and store in a desiccator until ready to count. Postpone counting for at least four hours to permit decay of short-lived natural radioactivity.

(6) Transfer the filtrate to a pyrex beaker and evaporate to near dryness without boiling (a gentle stream of filtered air may be passed into the beaker to speed up the evaporation).

(7) Transfer the residue to a weighed counting planchet with the aid of a wash bottle and a rubber or plastic policeman.

(8) Rinse the beaker with a few ml of distilled water (plus not more than 1 ml of 1 N HCl), police down the beaker

walls and transfer the rinse liquids to the counting planchet, dry (103°C), weigh, and store in a desiccator until ready to count.

b. Silt Samples

(1) Remove any extraneous matter, such as gravel or bits of vegetation, from a representative portion of the sample, and, if necessary, air dry and grind to a homogeneous powder.

(2) Make a thin slurry with distilled water and transfer enough sample to each of three counting planchets to give weights of about 50, 100, and 200 mg, respectively.

(3) Dry in an oven at 103°C, weigh (Note 1), and store in a desiccator until ready to count.

(4) The 50 and 100 mg samples are alpha-counted; the 100 and 200 mg samples are beta-counted.

c. Biological Materials

(1) Process a sample of 2 to 10 grams.

(2) Remove any visible extraneous matter, such as silt, from the specimen by washing with distilled water, blot dry, and re-weigh to determine live weight.

(3) Place the sample in a weighed porcelain or silica dish (or in a counting planchet), and determine dried solids (103°C) and fixed solids (600°C).

(4) Distribute up to 200 mg of fixed solids in a counting planchet.

(5) Dry (103°C), cool, weigh (Note 1), and store in a desiccator for counting (Note 2).

Counting Procedure

(1) Alpha counting: Count the sample at the alpha operating voltage for a time sufficient to give the required precision.

(2) Beta counting: Count the sample at the beta operating voltage for a time sufficient to give the required precision.

## Calculation

(1) General calculations applicable for the assaying of gross alpha or gross beta activity in any counter are as follows:

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)} = \text{gross radioactivity, } \mu\mu c/l \pm \text{C.E.}^{*}$$

where:

A = counting efficiency factor
B = self absorption factor
C = sample volume, liters

$$^{*}\text{C.E.}(\pm\ \mu\mu c/l) = \frac{1.96\sqrt{\frac{cpm_s}{tm} + \frac{cpm_b}{Tm}}}{(A)(B)(C)(2.22)}$$

where:

$cpm_s$ = counts per minute, sample
$cpm_b$ = counts per minute, background
tm = counting time, sample
Tm = counting time, background

## Notes

(1) If sample solids are powdery, treat them with 1 ml of a solution of lucite in acetone (0.5 grams per 100 ml), and enough additional acetone to wet the sample completely, re-dry, and count.

(2) Some biological materials, especially those containing fatty tissues, are difficult to process. An alternate method is to digest 2 to 10 grams of sample in a porcelain or silica dish with 6 N $HNO_3$ first at room temperature until liquefied and subsequently at a boiling temperature. Evaporate to dryness and ignite at 600°C. Cool, weigh, transfer known aliquots of up to 200 mg to counting planchets and proceed as described previously.

# IV. ISOTOPIC ANALYSES

## (RADIOCHEMICAL PROCEDURES)

# TRITIUM

## Principle of Method

The water sample is distilled to remove quenching materials or non-volatile radioactive materials. The distillation is carried to dryness to insure complete transfer of the tritium from the residue to the distillate. A portion of the distillate is mixed with scintillation solution, cooled, and counted in a liquid scintillation spectrometer (coincidence type). Standard tritium and background samples are prepared and counted simultaneously to nullify errors produced by aging of the scintillation medium or instrument drift.

## Equipment

Liquid scintillation spectrometer.

## Reagents

p-Dioxane, (1,4 - Dioxane).

Anisole, (Methoxybenzene).

1,2 - Dimethoxyethane (Ethylene glycol dimethyl ether).

PPO, (2, 5 - Diphenyloxazole).

POPOP, (1,4 - di [2 - (5 - phenyloxazolyl)] benzene).

Scintillation solution (freshly prepared). Mix thoroughly 6 parts 1, 4 - dioxane (by volume), 1 part anisole (by volume), 1 part dimethoxyethane (by volume). Add PPO, to contain 15.0 grams per liter and POPOP, to contain 0.62 grams per liter.

## Procedure

(1) Distill 30-40 ml of sample to dryness (Note 1).

(2) Pipette 16 ml of scintillation solution into a 5-dram scintillation vial.

(3) Pipette 4 ml of the sample distillate into the scintillation vial, cover tightly with screw cap and shake until thoroughly mixed.

(4) Prepare a background sample in the same manner as the sample consisting of 4 ml of distilled water and 16 ml of scintillation solution.

(5) Prepare a standard in the same manner as the sample consisting of 16 ml of scintillation solvent and 4 ml of distilled water containing a known amount of tritium activity.

(6) Cool the sample, background, and standard solutions in the instrument freezer to prepare for counting.

(7) In normal counting operation, tritium is counted with a window setting of approximately 10-100 volts. This roughly approximates the ideal window where the sample-rate squared divided by the background is a maximum. The high voltage is set so as to obtain the peak counting efficiency in the window. The sample is usually counted for 100 minutes to obtain good statistics, although this time may be adjusted as appropriate.

Calculation

$$\frac{A - B}{C} = \text{counting efficiency (D)}$$

$$\frac{E - B}{(D)(F)(G)} = \text{tritium, } \mu\mu c/ml$$

where:

A = gross cpm of standard

B = cpm background

C = dpm of tritium activity in standard

D = counting efficiency, cpm per dpm

E = gross cpm for sample

F = sample volume, (4 ml)

G = conversion factor, dpm/$\mu\mu$c (2.22)

Notes

(1) A large sample is distilled to dryness to reduce the possibility of loss of tritium.

Reference

1. Davidson, J. D., and Feigelson, P., "Practical Aspects of Internal Sample Liquid Scintillation Counting." The Internal Journal of Applied Radiation and Isotopes, 2 (1), 1-18 (1957).

V. J. S.
11-16-59

# COBALT AND ZINC

## Principle of Method

Cobalt and zinc are retained and concentrated on a cation exchange resin from a dilute HF solution. After elution from the cation resin with strong HCl, the resulting effluent solution is then passed over an anion exchange column where the cobalt and zinc are retained as anion complexes. Sequential elution separates the cobalt from the zinc after which each fraction is evaporated and counted.

## Reagents

Anion exchange resin, strong basic form ($Cl^-$) x 8, 50-100 mesh.

Cation exchange resin, strong acid form ($H^+$) x 8, 50-100 mesh.

Cobalt carrier, 20 mg/ml.

Hydrochloric acid, 12 N.

Hydrochloric acid, 1 N .

Hydrofluoric acid, 1 N .

Nitric acid, 1 N .

Zinc carrier, 20 mg Zn/ml.

## Procedure

(1) Add 1 ml each of zinc and cobalt carrier (Note 1) and 7 ml of 1.0 N hydrofluoric acid (Note 2) to a one liter sample.

(2) Pass solution through a 2 gram column (Note 3) of cation resin at a rate of 1 ml/$cm^2$/min (Note 4).

(3) Elute the cobalt and zinc from the cation resin with 50 ml 12 N HCl at the same rate as above.

(4) Pass the 50 ml of cation eluate through a 2 gram column (Note 3) of anion exchange resin, previously washed with concentrated HCl, discarding the effluent.

(5) Pass 25 ml of 1 N HCl over anion exchange resin at 1 ml/$cm^2$/min. This portion contains the cobalt.

(6) Evaporate to dryness in the beaker and transfer to a stainless steel planchet. Dry under infra-red lamp and beta count.

(7) Pass 75 ml of 1 N $HNO_3$ through the anion resin at 1 ml/$cm^2$/min, this portion contains the zinc. This is evaporated nearly to dryness, transferred to a stainless steel planchet, dried under infra-red lamp, and counted (Note 5).

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)} = \text{cobalt or zinc, } \mu\mu c/l$$

where:

A = efficiency factor (counting), cpm/dpm

B = chemical yield

C = volume, liters

## Notes

(1) Carriers are needed only to determine quantatively the percentage recovery.

(2) Addition of HF complexes Zr-Nb which then passes through the cation column.

(3) For samples of higher solids content, larger resin columns may be required.

(4) Recommended column sizes are 0.8 cm x 5 cm with flow rate of one drop each four seconds.

(5) If carriers are added, the cobalt and zinc eluates may each be neutralized with NaOH, centrifuged, and the cobalt or zinc transfered to a stainless steel planchet with dilute acid for counting.

## Reference

1. Krieger, H. L., Gilchrist, J., and Gold. S., "The Concentration of Radioactivity and the Detection of Cobalt-60 and Zinc-65 in Rainout". Paper presented at 3rd Conference on Analytical Chemistry in Nuclear Reactor Technology, Gatlinburg, Tenn., October, 1959.

H.L.K., J.E.G., S.G.
11-16-59

# RADIOSTRONTIUM AND RADIOBARIUM

## Principle of Method

Strontium and barium carriers are added to a water sample and precipitated as carbonates. Strontium and barium are partially separated from calcium by precipitation with strong nitric acid. Anhydrous acetone is used to separate the remaining calcium nitrate from strontium and barium nitrates. Rare earths and trivalent cations are removed by two hydroxide scavenges. Barium is separated from strontium as the chromate. Strontium is collected as the oxalate and measured for total strontium. Barium is converted from the chromate to the chloride for determination.

## Reagents

Acetone, anhydrous.

Ammonium acetate buffer, pH 5.0.

Alcohol, absolute.

Alcohol, 95 per cent.

Ammonium hydroxide, 15 N.

Ammonium hydroxide, 6 N.

Ammonium oxalate, saturated.

Barium carrier, 20 mg/ml.

Ethyl ether.

Hydrochloric acid, 12 N.

Hydrochloric acid - anhydrous ether solution, 5:1 ratio.

Nitric acid fuming 100 per cent.

Nitric acid, 16 N.

Nitric acid, 6 N.

Phenolphthalein indicator, 0.1 per cent.

Rare earth carrier, Mixed.

Sodium carbonate, 3 N.

Sodium chromate, 3 N.

Sodium hydroxide, 6 N.

Strontium carrier, 20 mg/ml.

Procedure

(1) Add 1 ml each of Sr and Ba carriers to one liter of water.

(2) Heat to boiling, add 20 ml of 6 N NaOH and 20 ml of 3 N $Na_2CO_3$, stir and digest for approximately 30 minutes, then set beaker aside until precipitate has settled (approximately 1-2 hr).

(3) Siphon or decant a major portion of the clear supernate liquid and separate the remaining precipitate and supernate by centrifugation in a 40 or 50 ml centrifuge tube.

(4) Dissolve the precipitate in 2 ml of 16 N $HNO_3$, adding dropwise, add 1 ml of $H_2O$, and heat to boiling.

(5) Add 25 ml of fuming $HNO_3$, cool in an ice bath with stirring and centrifuge, discarding the supernate. Invert the tube in a beaker, allowing the excess $HNO_3$ to drain off.

(6) Add 25 ml of anhydrous acetone, cool in an ice bath with stirring and centrifuge.

(7) Repeat step 6 if calcium nitrate is still present.

(8) Dissolve the precipitate in 3-5 ml $H_2O$ and heat to boiling to expel remaining acetone, add 15-20 ml water.

(9) Add 1 ml of mixed rare earth carrier and make solution basic with 6 N $NH_4OH$. Heat in a water bath for 5-10 minutes, centrifuge, and decant supernate to a clean tube.

(10) Repeat step 9.

(11) Add phenolphthlein indicator and neutralize solution with 6 N $HNO_3$, add 5 ml of ammonium acetate buffer. Heat to boiling and add 1 ml 3 N $Na_2CrO_4$, stir for a minute, and centrifuge, decanting the supernate into a clean tube.

(12) Add about 5 ml of 12 N HCl to the $BaCrO_4$ precipitate from the above step, heat in a water bath until the precipitate is dissolved and only a granular white precipitate of $BaCl_2$ remains. Cool in an ice bath and add 20 ml of cold HCl-ether mixture, stir well and centrifuge, saving the precipitate. Only a pure precipitate should remain, if not repeat the HCl-ether application.

(13) Transfer with absolute alcohol on a stainless steel planchet, dry, weigh, and count the $BaCl_2$ precipitate in an internal proportional counter.

(14) Add 2 ml of conc $NH_4OH$ to the supernatant from step 11, heat to boiling and add 5 ml of saturated ammonium oxalate. Stir, then centrifuge, discarding the supernate.

(15) Wash the precipitate twice with hot water, centrifuging after each wash. Wash 3 times with 95 per cent ethyl alcohol, then transfer the precipitate with ethyl alcohol to a stainless steel planchet, dry, weigh, and count the total strontium, $SrC_2O_4 \cdot H_2O$, in an internal proportional counter (Note 1).

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(2.22)} = \text{radiostrontium or radiobarium, } \mu\mu c/l$$

where:

A = efficiency factor

B = chemical yield

C = self absorption factor

D = volume, liters

## Notes

(1) If $Sr^{90}$ is also desired, the oxalate precipitate may be set aside for $Y^{90}$ ingrowth.

## References

1. Hahn, R. B. and Straub, C. P. "Determination of Radioactive Strontium and Barium in Water." Jour. Amer. Water Works Assn., 47 (4): 335 (April 1955).

C.P.S., R.B.H.
11-16-59

# STRONTIUM-90

## Principle of Method

Carrier strontium is added and precipitated as the carbonate. The precipitate is dissolved in hydrochloric acid and purified by hydroxide scavenging. The strontium is precipitated as the carbonate and allowed to stand for ingrowth of yttrium-90. The activity (yttrium-90) contained in this precipitate is extracted with equilibrated TBP, washed from the organic with 0.1 N $HNO_3$, dried in a stainless steel planchet, and counted in a low background beta anti-coincidence counter.

## Reagents

Ammonium acetate buffer, pH 5.0.

Ammonium hydroxide, 6 N.

Barium carrier, 20 mg/ml.

Cerium carrier, 10 mg/ml.

Hydrochloric acid, 6 N.

Iron carrier, 20 mg/ml.

Methyl orange, 0.1 per cent.

Nitric acid, fuming, 16 N, 14 N, 4 N, 0.1 N.

Sodium carbonate, 3 N.

Sodium chromate, 1.0 N.

Sodium hydroxide, 6 N.

Strontium carrier, 88 mg/ml.

Tributyl phosphate, (equilibrated with 14 N $HNO_3$).

Zirconium carrier, 10 mg/ml.

## Procedure

(1) Transfer a one liter aliquot of sample into a 1500 ml beaker and add 1.0 ml strontium nitrate carrier and 10 ml 6 N NaOH.

(2) Heat to boiling and add 5 ml 3 N $Na_2CO_3$.

(3) Allow the precipitate to settle and after one or two hours, decant, and discard the supernatant.

(4) Collect the precipitate in a 50 ml centrifuge tube.

(5) Dissolve the precipitate in 3 ml 6 N HCl.

(6) Add 1 ml barium carrier, 15 to 20 ml $H_2O$, and 5 drops methyl orange indicator (Note 1).

(7) Add 6 N $NH_4OH$ dropwise and with stirring, to a methyl orange end point.

(8) Add 1 ml 1.0 N $K_2CrO_4$ and warm in a water bath.

(9) Slowly add 5 ml of acetate buffer solution. Allow the precipitate to digest 5 to 10 minutes, and then add 1 ml barium carrier.

(10) Centrifuge and discard the $BaCrO_4$ precipitate.

(11) Make the supernate alkaline with 6 N $NH_4OH$ and add 5 ml of 3 N $Na_2CO_3$.

(12) Centrifuge, discard the supernatant, and wash the precipitate with water containing a few drops of 3 N $Na_2CO_3$ and discard the washings.

(13) Dissolve the $SrCO_3$ precipitate in 3 of 6 N HCl.

(14) Add 20 to 25 ml of $H_2O$, 1 ml iron carrier, heat and make alkaline with 6 N $NH_4OH$.

(15) Add 1 ml iron carrier, centrifuge, and discard the precipitate.

(16) Add to the supernatant 1 ml zirconium carrier and 1 ml cerium carrier.

(17) Add 6 N HCl to dissolve any precipitate, warm and make alkaline with 6 N $NH_4OH$.

(18) Add 1 ml each of zirconium and cerium carriers, centrifuge, and discard the precipitate. Record time of precipitation (Note 2).

(19) Add sufficient $Na_2CO_3$ to the supernatant for complete precipitation of strontium carbonate.

(20) Store this precipitate for two weeks (Note 3).

(21) Dissolve the precipitate in 2 ml 4 N $HNO_3$.

(22) Cool in an ice bath and add with stirring 10 ml of fuming nitric acid.

(23) Centrifuge and transfer the supernate into a 60 ml separatory funnel.

(24) Dissolve the residue in 2 ml 4 N $HNO_3$ and repeat steps 22 and 23.

(25) Add 6 ml 4 N $HNO_3$ to the separatory funnel.

(26) Add 5.0 ml equilibrated TBP and shake thoroughly for 10 minutes (Note 4).

(27) Transfer the organic layer to another separatory funnel and repeat step 26 twice.

(28) Wash combined organic phases three times with 5 ml portions 14 N $HNO_3$. Discard the washings.

(29) Back extract the combined organic phases with 10 ml 0.1 N $HNO_3$ for 10 minutes.

(30) Transfer the aqueous phase to a 50 ml beaker and reduce its volume on a hot plate.

(31) Repeat steps 29 and 30 twice, combining the aqueous phases in the 50 ml beaker.

(32) Evaporate the solution to about 10 ml and transfer it into a stainless steel planchet and evaporate to dryness.

(33) Wash beaker several times with small volumes (2 ml) of 0.1 N $HNO_3$, transferring each wash to the planchet.

(34) Determine the activity in a low background beta (anti-coincidence) counter.

Calculation.

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(F)(2.22)} = \text{Strontium-90, } \mu\mu c/l \pm (C.E)^{*}$$

where:

A = Efficiency factor for stainless steel in low background (.396)

B = per cent extraction factor (0.98)

C = per cent ingrowth factor (0.97)

D = chemical yield (Note 5)

E = sample volume, liters

F = decay factor; calculated from the following formula:

$$A = A_0 e^{-\lambda t}$$

where:

A = activity remaining after a time interval, t

$A_o$ = activity of sample at some original time

$$\lambda = \frac{0.693}{\text{half life of yttrium (64.2 hrs)}}$$

t = time (hours) from separation to counting

Notes

(1) Barium removal is done in step 6 through step 12. These steps may be omitted unless total radiostrontium is to be determined subsequently or the $Ba^{140}$ content is expected to be very high.

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$$\text{C.E., } \mu\mu c/l = \frac{1.96\sqrt{\frac{cpm_s}{t_m} + \frac{cpm_b}{T_m}}}{(A)(B)(C)(D)(E)(F)(2.22)}$$

(2) This is start of ingrowth time for yttrium-90.

(3) The strontium is left in solid form as $SrCO_3$ so that the yttrium-90 formed may be trapped in the crystal, thus minimizing adsorption losses to glassware, etc.

(4) This is the end of ingrowth time for yttrium-90; start of decay time of yttrium-90.

(5) Chemical yield is determined by flame photometry.

Reference

1. Goldin, A. S., Velten, R. J., and Frishkorn, G. W., "Determination of Radioactive Strontium", Anal. Chem., 31, 1940 (1959).

A.S.G., R.J.V.
11-16-59

# RADIOSTRONTIUM

## Principle of Method

After separation of yttrium from strontium by nitrate precipitation, strontium nitrate is reprecipitated as the carbonate and total strontium activity is determined. Strontium-89 is determined by difference.

## Reagents

Ammonium hydroxide, 6 N.

Sodium carbonate, 3 N.

## Procedure

(1) Dissolve the residue from step 24 (38B-1) with 15 ml distilled water.

(2) Make the solution alkaline with 6 N $NH_4OH$.

(3) Add sufficient 3 N $Na_2CO_3$ slowly for complete precipitation of strontium carbonate.

(4) Centrifuge, check for completeness of precipitation, decant and discard the supernate.

(5) Slurry, with the aid of a wash bottle, and transfer the precipitate into an aluminum planchet.

(6) Dry precipitate in oven at 105°-110°C.

(7) Cool, place the planchet in an internal proportional counter, and determine the beta activity.

## Calculation

$$\frac{\text{Net cpm} - (E)}{(A)(B)(C)(D)(2.22)} = \text{strontium-89, } \mu\mu c/l$$

where:

A = efficiency factor for aluminum

B = chemical yield

C = self absorption factor for strontium-89

D = volume, liters

E = net cpm due to strontium-90 (Note 1)

**Notes**

(1) The cpm of strontium-90 is calculated from the total dpm from procedure 38B, after applying self absorption correction and counter detection efficiency for an internal porportional counter.

**Reference**

1. Goldin, A. S., Velten, R. J., and Frishkorn, G. W. "Determination of Radioactive Strontium." Anal. Chem., 31, 1940 (1959).

# ZIRCONIUM-NIOBIUM

## Principle of Method

In an aqueous solution zirconium-niobium is easily complexed by a very dilute solution of hydrofluoric acid. These anion complexes are concentrated by passage through anion exchange resin, and then sequentially eluted from the anion resin with a HCl-HF mixture and an $HNO_3$ solution (Note 1).

## Reagents

Anion exchange resin, strong basic form, ($Cl^-$) x 8, 50-100 mesh.

Cation exchange resin, strong acid form, ($H^+$) x 8, 50-100 mesh.

Cupferron, 6 per cent.

Hydrochloric-hydrofluoric acid mixture, 8 N HCl, 0.3 N HF.

Hydrofluoric acid, 1.0 N.

Niobium carrier, 20 mg/ml.

Nitric acid, 1 N.

Zirconium carrier, 10 mg/ml.

## Procedure

(1) Add 7.0 ml 1.0 N HF to one liter water sample (Note 2).

(2) Pass through cation and anion exchange resin columns in series at a flow rate of 1 ml/$cm^2$/min (Note 3)(Note 4).

(3) Pass 10-15 ml of ether through the anion column after as much water has been drained off as possible and pass air through the column to evaporate the ether.

(4) Pass 20 ml of 8 N HCl - 0.3 N HF mixture through the column (1 drop/6 sec). This portion contains the zirconium. Precipitate with cupferron, filter, ignite to $ZrO_2$, weigh and count in an internal proportional counter.

(5) Elute the niobium from the column with 75 ml of 1 N $HNO_3$. Neutralize with $NH_4OH$, filter, ignite to $Nb_2O_5$, weigh, and beta count.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(2.22)} = \text{zirconium or niobium, } \mu\mu c/l$$

where:

A = efficiency factor

B = chemical yield

C = self absorption factor

D = volume, liters

## Notes

(1) This procedure may be used in conjunction with the cobalt and zinc procedure, as the cobalt, zinc, and other cations will be retained on the cation resin preceeding the anion resin where the zirconium-niobium fluoride complexes are retained.

Ruthenium, if present, will be a radioactive contaminent of zirconium and niobium and must be first separated radiochemically.

(2) Zirconium and niobium carriers may be added at this point if it is necessary to determine percentage recovery. For water samples the zirconium-niobium recovery should be above 95%.

(3) Recommended column size is 0.8 cm x 5 cm length with a flow rate of 1 drop each 4 seconds.

(4) The cation resin column is necessary, as $CeF_3$ will be partially retained by an anion exchange column.

## Reference

1. Krieger, H. L., Gilchrist, J., Gold, S., "The Concentration of Radioactivity and the Detection of Cobalt-60 and Zinc-65 in Rainout." Paper presented at 3rd Conference on Analytical Chemistry in Nuclear Reactor Technology, Gatlinburg, Tenn., October, 1959.

H.L.K., J.E.G., S.G.
11-16-59

# RADIOIODINE

## Principle of Method

Carrier sodium iodide is oxidized by sodium hypochlorite in basic solution to iodate and periodate. It is then reduced to iodide by sodium bisulfite in acid solution. This procedure assures interchange of carrier and radioiodine. Sodium nitrite is added to oxidize to elemental iodine, which is extracted into carbon tetrachloride. This serves as both a purification and concentration step. The iodine is further purified and concentrated by back-extraction into sodium bisulfite solution which is finally gamma-counted at the I-131 photo peak.

## Reagents

Carbon tetrachloride, reagent grade.

Hydrochloric acid, 6 N.

Iodine carrier, 13 mg/ml.

Sodium bisulfite, 1 N.

Sodium hydroxide, 6 N..

Sodium hypochlorite,.5.25 per cent. (Note 1.)

Sodium nitrite, 1 N.

## Procedure

(1) Transfer a 500 ml aliquot of sample into a one liter beaker.

(2) Add 1.0 ml of iodine carrier , 5 ml 6 N NaOH, 1 ml of NaClO and heat to near boiling.

(3) Cool and transfer the solution to a separatory funnel and add 10 ml 6 N HCl (Note 2).

(4) Add sufficient 1 N $NaHSO_3$ dropwise to reduce all of the iodate to iodide.

(5) Add 5 ml $CCl_4$ and 3 ml 1 N $NaNO_2$. Shake thoroughly and draw off the organic layer into a 60 ml separatory funnel containing 2 ml 1 N $NaHSO_3$ solution.

(6) Repeat step 5 using 1 ml 1 N $NaNO_2$.

(7) Repeat step 6 twice and discard the aqueous layer (Note 3).

(8) Shake separatory funnel thoroughly until $CCl_4$ layer is decolorized, allow phases to separate, and transfer aqueous layer to a counting tube (Note 4).

(9) Add 2 ml 1 N $NaHSO_3$ to separatory funnel and repeat step 8.

(10) Add 1 ml $H_2O$ to the separatory funnel and repeat step 8.

(11) Count activity contained in combined aqueous extracts in a well-type gamma scintillation counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)} = \text{Iodine-131, } \mu\mu c/l.$$

where:

A = counting efficiency
B = chemical yield
C = volume, liters

## Notes

(1) The concentration of this reagent can be checked by standard methods such as arsenious oxide titration method. (A.O.A.C., 6.139, 1945).

(2) The pH of the solution should be less than 2.0. The yellow or brown color of $I_2$ should not be present.

(3) A total of four extractions should suffice for adequate removal of iodine into the organic layer. The color of the iodine-carbon tetrachloride layer may be used to judge the degree of extraction.

(4) A 10 ml or 20 ml lusteroid centrifuge tube or suitable glass or plastic vial may be used.

## Reference

1. Glendenin, L. E., and Metcalf, R. P., Paper 278 in "Radiochemical Studies: The Fission Products," Coryell, C. D., and Sugarman, N., (eds), p. 1625, McGraw-Hill, New York, 1951.

# RADIOCESIUM

## Principle of Method

Cesium is recovered by precipitation as the ammonium phosphomolybdate, followed by purification, and final precipitation as the cobaltinitrite. The precipitate of cesium cobaltinitrite is counted for beta activity in an internal proportional counter.

## Reagents

Acetic acid, 18 N.

Ammonium hydroxide, 15 N.

Ammonium molybdate, 0.5 N.

Cesium carrier, 20 mg/ml.

Nitric acid, 16 N.

Phosphoric acid, 85 per cent.

Sodium nitrite, reagent grade.

Sodium cobaltinitrite reagent.

## Procedure

(1) Transfer a one liter aliquot of sample into a 1500 ml beaker.

(2) Acidify the solution with 50 ml 16 N $HNO_3$ and add 1.0 ml of cesium carrier.

(3) Add 0.5 ml $H_3PO_4$ and heat the solution to a temperature of 50°C.

(4) Add 50 ml 10 per cent ammonium molybdate and stir vigorously with the aid of a magnetic stirrer for about thirty minutes.

(5) Allow the precipitate of cesium ammonium phosphomolybdate to settle.

(6) Siphon or decant the bulk of the supernatant and discard it.

(7) Collect the precipitate in a fine fritted disc filter funnel and discard the filtrate.

(8) Place a 50 ml centrifuge tube under the funnel and add 5 to 10 ml of 6 N NaOH to the funnel. Allow the NaOH several minutes to dissolve the precipitate.

(9) Draw the solution into the centrifuge tube with the aid of suction.

(10) Wash the filter bed with a little water and suck the washings into the centrifuge tube.

(11) Add 5 ml acetic acid, with stirring, and add 2 grams sodium nitrite.

(12) Digest in a water bath for about 10 minutes.

(13) Cool in an ice bath, add 2.0 ml sodium cobaltinitrite reagent to precipitate cesium cobaltinitrite.

(14) Wash the precipitate with 10 ml distilled water at 10°C.

(15) Transfer the precipitate to an aluminum planchet. Dry at 100°C. and count the activity in an internal proportional counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)(1.08)} = \text{radiocesium, } \mu\mu c/l$$

where:

A = efficiency factor

B = self absorption factor

C = volume, liters

## Reference

1. Kahn, B., Smith, D. K., and Straub, C. P., Anal. Chem., 29, 1210, August (1957).

A.S.G., G.W.F.
11-16-59

# TOTAL RARE EARTHS

## Principle of Method

Carrier cerium (representing all the rare earth activities) is added and precipitated as the fluoride. The precipitate is dissolved in boric acid and nitric acid and precipitated as the hydroxide. Zirconium activity is removed from the precipitate by iodate precipitation after reducing all rare earths to the trivalent state. After subsequent hydroxide scavenging to remove alkaline earths, the total rare earths are precipitated as oxalates and beta-counted in an internal proportional counter.

## Reagents

Ammonium hydroxide, 15 N.

Barium carrier, 20 mg/ml.

Boric acid, saturated.

Cerium carrier, 20 mg/ml.

Ethyl alcohol, 95 per cent.

Hydrochloric acid, 12 N, 6 N.

Hydrofluoric acid, 48 per cent.

Hydrogen peroxide, 30 per cent.

Iodic acid, 0.35 N.

Nitric acid, 16 N.

Oxalic acid, saturated.

Sodium carbonate, 3 N.

Sodium hydroxide, 19 N.

Strontium carrier, 20 mg/ml.

Zirconium carrier, 10 mg/ml.

**Procedure**

(1) Add 1 ml cerium carrier to one liter of water. stir, and heat to boiling.

(2) Add 2 ml 19 N NaOH, 10 ml 3 N $Na_2CO_3$, stir, and heat for 30 minutes. Settle until supernate is clear.

(3) Decant supernate and transfer precipitate to a 50 ml lusteroid tube. Centrifuge and discard supernate.

(4) Wash precipitate with 30 ml distilled water.

(5) Dissolve precipitate with 1 drop $H_2O_2$ and 5 ml 6 N HCl. Add 1 ml zirconium holdback carrier and precipitate fluorides with 2 ml HF. Stir, centrifuge and decant.

(6) Wash precipitate with 20 ml distilled water.

(7) Dissolve precipitate in 2 ml saturated $H_3BO_3$ and 5 ml 6 N $HNO_3$. Transfer to glass centrifuge tube with a little distilled water.

(8) Dilute to 20 ml, add 2 ml 19 N NaOH to precipitate cerium hydroxide. Stir, centrifuge, and discard supernate.

(9) Wash precipitate with 20 ml distilled water.

(10) Dissolve precipitate with 8 ml concentrated $HNO_3$, add 1 drop $H_2O_2$ and 0.5 ml zirconium carrier.

(11) Precipitate with 15 ml 0.35 N $HIO_3$, stir, centrifuge and transfer supernate to another glass centrifuge tube.

(12) Add 10 ml 19 N NaOH, stir, centrifuge, and discard supernate.

(13) Wash precipitate with 10 ml $H_2O$.

(14) Dissolve precipitate with 1 ml 6 N HCl. Add 1 ml each barium and strontium carriers and 15 ml $H_2O$.

(15) Add 1 ml 15 N $NH_4OH$, stir, centrifuge, and discard supernate.

(16) Dissolve precipitate with 1 ml 6 N HCl, add 15 ml $H_2O$ and 1 ml concentrated $NH_4OH$. Stir, centrifuge and discard supernate.

(17) Wash precipitate twice with 10 ml $H_2O$.

(18) Dissolve precipitate with 1 ml 6 N HCl, add 15 ml $H_2O$ and slowly bring to a boil. Add 15 ml saturated $H_2C_2O_4$ and cool in ice bath for 10 minutes.

(19) Centrifuge, wash precipitate with 10 ml hot water.

(20) Transfer to tared planchet with 3 portions of ethyl alcohol and dry under infra-red lamp.

(21) Weigh and count in internal proportional counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)} = \text{Total rare earths, } \mu\mu c/l$$

where:

A = efficiency factor

B = chemical yield

C = volume, liters

## Reference

1. Boldridge, W. F. and Hume, D. N., Paper 294 in "Radiochemical Studies: The Fission Products," Coryell, C. D., and Sugarman, N., (eds), p. 1693, McGraw-Hill, New York, 1951.

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11-16-59

# RADIUM-226

## Principle of Method

Radium is separated by precipitation on barium sulfate, purified by solution, and reprecipitated in an EDTA cycle. The precipitate is counted for alpha activity in an internal proportional counter.

## Reagents

Acetic acid, 18 N.

Ammonium hydroxide, 15 N, 6 N.

Barium carrier, 6.7 mg/ml.

Citric acid, 3 N.

Ethylenediaminetetraacetic acid, (EDTA) M/4.

Lead carrier, 100 mg/ml.

Methyl orange indicator, 0.1 per cent.

Nitric acid, 16 N.

Sulfuric acid, 18 N.

## Procedure

(1) Transfer a one liter aliquot of sample into a 1500 ml beaker (Note 1).

(2) Add 5 ml 3 N citric acid, 2.5 ml 15 N $NH_4OH$, 2 ml lead carrier, and 1.0 ml barium carrier.

(3) Heat the solution to boiling and add to the hot solution 10 drops of methyl orange indicator.

(4) Add with stirring, sulfuric acid to a pink color and then 0.25 ml in excess.

(5) Digest 5 - 10 minutes, allow the precipitate to settle 1 to 2 hours, decant, and discard the supernatant.

(6) Collect the precipitate in a 50 ml centrifuge tube.

(7) Wash the precipitate twice with 10 ml portions of $HNO_3$, discarding the washings.

(8) Dissolve the precipitate in 10 ml water, 10 ml EDTA, and 3 ml 6 N $NH_4OH$.

(9) Warm the solution, add 2 ml glacial acetic acid dropwise and digest 5 - 10 minutes. Centrifuge, discard the supernatant and record time (Notes 2 and 3).

(10) Wash the reprecipitated $BaSO_4$ with water and transfer it into a centrifuge tube adapted for centrifuging precipitates onto planchets (Note 4).

(11) Allow the precipitate to settle 5 to 10 minutes, centrifuge, dry, flame, weigh, and determine the alpha activity in an internal proportional counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(2.22)} = \text{radium-226, } \mu\mu c/l$$

where:

A = ingrowth factor (Note 5)

B = efficiency factor for alpha counting

C = self absorption factor

D = chemical yield

E = volume, liters

## Notes

(1) Larger samples may be taken if desired, with suitable modification of the quantity of reagents used in concentration. The quantity of barium carrier, however, should not be increased.

(2) This quantity of acetic acid is in approximately 2:1 excess over the $NH_4OH$, giving a pH of about 4.5. This is sufficient to destroy the Ba-EDTA complex, but not the Pb-EDTA.

(3) From this time on, Rn (and daughters) grow into the $BaSO_4$ precipitate.

(4) Atomlab-Eckstein centrifuge tube. If not available, the precipitate can be transferred in any suitable manner.

(5) Can be calculated from data of Kirby, in Atomic Energy Commission Report MLM-859. Part of this data is reproduced in Table I.

Table I

Growth of Alpha and Beta Activities from Initially Pure Parents during the First 120 Hours

| Parent: Hours | Radium-226 $at/N_o \lambda_1$ |
|---|---|
| 0 | 1.0000 |
| 1 | 1.0160 |
| 2 | 1.0363 |
| 3 | 1.0580 |
| 4 | 1.0798 |
| 5 | 1.1021 |
| 6 | 1.1238 |
| 24 | 1.4892 |
| 48 | 1.9054 |
| 72 | 2.2525 |

Reference

1. "Analysis of Water for Dissolved Radium-226," Goldin, A. S., In Charge Radiochemical Methods, Radiological Health Program, Robert A. Taft Sanitary Engineering Center, U. S. Public Health Service, Cincinnati 26, Ohio.

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11-16-59

# V. SPECIAL PROCEDURES APPLICABLE TO SPECIFIC SAMPLE TYPES

# DETERMINATION OF COBALT AND ZINC IN BIOLOGICAL MATERIALS

## Principle of Method

Cobalt and zinc in biota may be determined by the same anion exchange procedure used for cobalt and zinc in water, by first boiling with concentrated nitric acid, adding perchloric acid, and evaporating to near dryness. After leaching with dilute hydrochloric acid (Note 1) and evaporating to dryness, the cobalt and zinc are taken up in concentrated hydrochloric acid and passed through an anion exchange resin where they are quantitatively retained as chloride complexes. Other non-complexing cations do not interfere since they pass into the effluent. The cobalt and zinc are sequentially eluted from the resin with hydrochloric and nitric acids.

## Reagents

Anion exchange resin, strong basic form ($Cl^-$) x 8, 50-100 mesh.

Cobalt carrier, 20 mg/ml.

Hydrochloric acid, 12 N, 1 N.

Niobium carrier, 10 mg/ml.

Nitric acid, 16 N, 1 N.

Perchloric acid, 70 per cent.

Zinc carrier, 20 mg/ml.

Zirconium carrier, 10 mg/ml.

## Procedure

(1) Add to sample (Note 2) 1 ml each of the carrier cobalt, zinc, zirconium, and niobium.

(2) Add 50 ml 16 N $HNO_3$ and evaporate to approximately 25 ml.

(3) Add 15 ml 70 per cent $HClO_4$ and continue evaporation to near dryness (3-5 ml).

(4) Add 25 ml 1 N HCl heat to near boiling and slurry into a centrifuge tube, spin and decant supernate into clean beaker.

(5) Repeat step 4 twice using 10-15 ml of 1 N HCl.

(6) Evaporate the 1 N HCl solution to dryness and add 25-30 ml of concentrated HCl.

(7) Pass the concentrated HCl solution through a treated anion exchange resin column (Note 3) at a slow rate, then pass 5 ml of concentrated HCl through the column to remove remaining solution from the resin.

(8) Proceed with cobalt and zinc determination (RC-27A-1), step 3.

Notes

(1) Zirconium and niobium, which normally would interfere with the cobalt-zinc anion exchange determinations, are insoluble as the phosphates and are not leached by dilute acid. In small biological samples the concentration of phosphate might not be sufficient to precipitate the carrier zirconium-niobium, thus in such an instance, 1 ml of 1 N phosphoric acid should be added.

(2) Sample sizes should be about 20 grams moist weight. Volumes of acids used for wet ashing should be varied in accordance with sample size taken.

(3) A small column 2 grams (moist weight) or ~ 1 x 5 cm is sufficient. Pretreatment of the column consists of having concentrated HCl on the resin before passage of the sample solution. A slow rate is about 1 drop/6-8 seconds.

Reference

1. See (RC-27A-1).

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# DETERMINATION OF STRONTIUM-90 IN RIVER WATER

## Principle of Method

Carrier strontium is added and precipitated as the carbonate. The precipitate is dissolved in nitric acid and purified by hydroxide scavenging. The strontium is then precipitated from the supernatant solution with carbonate and allowed to stand for ingrowth of yttrium-90. The precipitate is then dissolved in nitric acid and the yttrium-90 is extracted with equilibrated TBP, washed from the organic with 0.1 N $HNO_3$, dried in a stainless steel planchet and counted in a low background beta anti-coincidence counter.

## Reagents

Ammonium hydroxide, 15 N, 6 N.

Hydrochloric acid, 6 N.

Nitric acid, fuming, 16 N, 14 N, 8 N, 4 N, 0.1 N.

Perchloric acid, 70-72 per cent.

Rare earth carrier, mixed.

Sodium carbonate, 3 N.

Sodium hydroxide, 6 N.

Strontium carrier, 88 mg/ml.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

## Procedure

(1) Transfer a 2.5-liter aliquot of sample into a three liter beaker and add 1.0 ml of strontium carrier.

(2) Heat to boiling on a hot plate and add 10 ml 6 N NaOH.

(3) Continue to heat and slowly, with stirring, add 20 ml 3 N $Na_2CO_3$.

(4) Allow the precipitate to settle at least two hours but preferably overnight.

(5) Siphon off the supernatant and discard it.

(6) Collect the precipitate in a 50-ml centrifuge tube. (Note 1).

(7) Carefully add 10 to 20 ml 8 N $HNO_3$ to the precipitate and transfer the contents into a 250-ml beaker. Rinse the centrifuge tube with 20 ml 16 N $HNO_3$ and add it to the beaker (Note 2).

(8) Evaporate to near dryness.

(9) Add 20 ml 16 N $HNO_3$ and again evaporate to near dryness.

(10) Add 20 ml 16 N $HNO_3$ and 10 ml of 70 per cent $HClO_4$ and heat to strong perchloric acid fumes.

(11) Cool, and add 20 ml of distilled water and transfer the contents into a 50-ml centrifuge tube.

(12) Centrifuge and decant the supernatant into another 50-ml centrifuge tube. Discard the residue.

(13) Make the solution alkaline with 15 N $NH_4OH$ to precipitate the insoluble hydroxides. Stir and centrifuge, transferring the supernatant into a clean 50-ml centrifuge tube.

(14) Dissolve the hydroxide precipitate in 2 to 3 ml 6 N HCl and repeat step 13 twice combining the supernates in the same centrifuge tube. Discard the final precipitate (Note 3).

(15) To this combined supernate add 1.0 ml of mixed rare earth carrier and digest in a hot water bath for 10 minutes.

(16) Centrifuge and decant the supernatant into a clean 50-ml centrifuge tube.

(17) Repeat steps 15 and 16 once (Note 4).

(18) Add sufficient $Na_2CO_3$ for complete precipitation of strontium carbonate (Note 5).

(19) Store this precipitate for two weeks.

(20) Dissolve the precipitate in 2 ml N $HNO_3$.

(21) Cool in an ice bath and add with stirring 10 ml of fuming nitric acid.

(22) Centrifuge and transfer the supernate into a 60-ml separatory funnel.

(23) Dissolve the residue in 2 ml 4 N $HNO_3$ and repeat steps 21 and 22 (Note 6).

(24) Add 6 ml 4 N $HNO_3$ to the separatory funnel to bring the aqueous phase to 30 ml.

(25) Add 5.0 ml of equilibrated TBP and shake thoroughly for 10 minutes.

(26) Transfer the organic layer to another 60-ml separatory funnel and repeat step 25 twice.

(27) Wash combined organic phases three times with 5 ml portions 14 N $HNO_3$.

(28) Back extract the combined organic phases with 10 ml 0.1 N $HNO_3$ for 10 minutes.

(29) Transfer the aqueous phase to a 50-ml beaker and reduce its volume on a hot plate.

(30) Repeat steps 28 and 29 twice, combining the aqueous phases in the 50-ml beaker.

(31) Evaporate the solution to about 10 ml and transfer it into a stainless steel planchet and evaporate to dryness.

(32) Wash beaker with small portions of 0.1 N $HNO_3$.

(33) Determine the activity in a low background beta (anti-coincidence) counter.

Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(F)(2.22)} = \text{Strontium-90, } \mu\mu c/l$$

where:

A = efficiency factor for stainless steel

B = per cent extraction factor (0.98).

C = per cent ingrowth factor (0.97).

D = chemical yield (Note 7)

E = volume, liters

F = decay factor, calculated from the following formula:

$$A = A_0 e^{-\lambda t}$$

where:

$A$ = activity remaining after a time interval, t.

$A_0$ = activity of sample at some original time

$\lambda = \frac{0.693}{\text{half life of yttrium (64.2 hrs.)}}$

$t$ = time (hours) from separation to counting

Notes

(1) If the precipitate is quite large, because of a high calcium content, it is more convenient to use a 250-ml centrifuge bottle.

(2) If there are no organics present, steps 8, 9, and 10 may be omitted.

(3) This precipitate contains insoluble hydroxides such as Al, Fe, Zr, and Nb. A subsequent scavenge of Zr, Fe, and Ce is performed in step 15.

(4) The rare earth scavenge is carried out to insure the separation of strontium and yttrium.

(5) This is the start of ingrowth time for Y-90.

(6) This is the end of ingrowth time for Y-90; start of decay time of Y-90.

(7) Chemical yield is determined gravimetrically or by flame photometry.

Reference

1. Goldin, A. S., Velten, R. J., and Frishkorn, G. W., "Determination of Radioactive Strontium," Anal. Chem., 31, 1940 (1959).

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# DETERMINATION OF STRONTIUM-90 IN SALINE WATER

## Principle of Method

Carrier strontium is added and magnesium is removed by precipitation with 8-quinolinol. Strontium is then precipitated as the carbonate, converted to the nitrate and purified by hydroxide scavenging. The strontium is stored as the carbonate for ingrowth of yttrium-90. The activity (yttrium-90) is extracted with equilibrated TBP, stripped with 0.1 N $HNO_3$, dried, and counted in a low background beta anti-coincidence counter.

## Reagents

Ammonium hydroxide, 15 N, 6 N.

Hydrochloric acid, 6 N.

Nitric acid, fuming, 16 N, 4 N, 0.1 N.

8-quinolinol solution, 7.5 per cent in methanol.

Rare earth carrier, mixed.

Sodium carbonate, 3 N.

Sodium hydroxide, 6 N.

Strontium carrier, 88 mg/ml.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

## Procedure

(1) Transfer a 1 liter sample into a 1500 ml beaker.

(2) Add 1.0 ml of strontium nitrate carrier.

(3) Heat to near boiling and add 10 ml 6 N $NH_4OH$.

(4) Add with stirring 100 ml of 8-quinolinol solution and allow the precipitate to settle at least 15 minutes.

(5) With the aid of a Buchner funnel filter through a double layer of Whatman #42 filter paper.

(6) Wash the precipitate with three 50 ml aliquots of distilled water.

(7) Transfer the filtrate to a 1500 ml beaker and add 30 ml 6 N NaOH. Heat to near boiling.

(8) Slowly add with stirring 50 ml 3 N $Na_2CO_3$.

(9) Allow the precipitate to settle and decant and discard most of the supernatant.

(10) Transfer the precipitate into a 250 ml centrifuge bottle, centrifuge and discard the supernatant.

(11) Wash the precipitate with 100 ml $H_2O$; centrifuge and decant the supernatant.

(12) Place the centrifuge bottle and precipitate in an oven at a temperature of 105°C. Allow the precipitate to dry overnight.

(13) Remove the centrifuge bottle from the oven, allow the precipitate to attain room temperature, and break up the precipitate with a stirring rod.

(14) Cautiously add with stirring 30 ml 16 N $HNO_3$ in 10 ml aliquots.

(15) Place the centrifuge bottle in a water bath and digest with occasional stirring.

(16) Cool in an ice bath with occasional stirring.

(17) Centrifuge and discard the supernatant.

(18) Add 30 ml 16 N $HNO_3$ and repeat steps 15 through 17.

(19) Dissolve the precipitate in 5 ml $H_2O$ and transfer the solution into a 50 ml centrifuge tube.

(20) Rinse the centrifuge bottle with two additional 5 ml aliquots of $H_2O$ and transfer the washings to the centrifuge tube.

(21) Add 1.0 ml of rare earth carrier and add with stirring a few drops of 15 N $NH_4OH$ until alkaline. Digest in a hot water bath for 10 minutes.

(22) Centrifuge and decant the supernatant into a clean 50 ml centrifuge tube.

(23) Repeat steps 21 and 22 once (Note 1).

(24) Add sufficient 3 N $Na_2CO_3$ for complete precipitation of strontium carbonate (Note 2).

(25) Store this precipitate for two weeks.

(26) Dissolve the precipitate in 2 ml of 4 N $HNO_3$.

(27) Cool in an ice bath and add with stirring 10 ml of fuming nitric acid.

(28) Centrifuge and transfer the supernate into a 60 ml separatory funnel.

(29) Dissolve the residue in 2 ml 4 N $HNO_3$ and repeat steps 27 and 28 (Note 3).

(30) Add 6 ml 4 N $HNO_3$ to the separatory funnel to bring the aqueous phase to 30 ml.

(31) Add 5.0 ml of equilibrated TBP and shake thoroughly for 10 minutes.

(32) Transfer the organic layer to another 60 ml separatory funnel and repeat step 31 twice.

(33) Wash the combined organic phases three times with 5 ml aliquots 14 N $HNO_3$.

(34) Back extract the combined organic phases with 10 ml 0.1 N $HNO_3$ for 10 minutes.

(35) Transfer the aqueous phase to a 50 ml beaker and reduce its volume on a hot plate.

(36) Repeat steps 34 and 35 twice, combining the aqueous phases in the 50 ml beaker.

(37) Evaporate the solution to about 10 ml and transfer it into a stainless steel planchet, and evaporate to dryness.

(38) Rinse the beaker with small portions 0.1 N $HNO_3$ and transfer the washings to the planchet. Evaporate to dryness.

(39) Determine the activity in a low background beta (anti-coincidence) counter.

Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(F)(2.22)} = \text{Strontium-90, } \mu\mu c/l$$

where:

A = efficiency factor on stainless steel

B = per cent extraction factor (0.98)

C = per cent ingrowth factor (0.97)

D = chemical yield (Note 4)

E = sample volume, liters

F = decay factor; calculated from the following formula:

$$A = A_0 e^{-\lambda t}$$

where:

A = activity remaining after a time interval, t

$A_0$ = activity of sample at some original time

$$\lambda = \frac{0.693}{\text{half life of yttrium (64.2 hrs)}}$$

t = time (hours) from separation to counting

Notes

(1) The rare earth scavenge is carried out to insure the separation of strontium and yttrium.

(2) This is the start of ingrowth time for Y-90.

(3) This is the end of ingrowth time for Y-90; start of decay time of Y-90.

(4) Chemical yield is determined gravimetrically or by flame photometry from residue of step 29.

Reference

1. Goldin, A. S., Velten, R. J., and Frishkorn, G. W., "Determination of Radioactive Strontium," Anal. Chem., 31, 1940 (1959).

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# DETERMINATION OF STRONTIUM-90 IN MILK BY TRICHLOROACETIC ACID PRECIPITATION

## Principle of Method

After addition of carrier strontium, the milk proteins are precipitated with trichloroacetic acid. Following filtration, excess oxalic acid is added and the alkaline earths are precipitated as the oxalates at pH 2.5 - 3.0, washed and ashed to the carbonates and/or oxides. The ash is dissolved in nitric acid and evaporated to dryness. The strontium is separated from calcium by repeated nitric acid treatment. The strontium nitrate is dissolved in water, scavenged by ferric iron and rare earth hydroxide precipitations, and stored as the carbonate for $Y^{90}$ ingrowth for two weeks. The $Y^{90}$ is separated with nitric acid, extracted into tributyl phosphate, stripped from the organic with dilute nitric acid, evaporated to dryness in a stainless steel planchet, and counted.

## Reagents

Ammonium hydroxide, 15 N, 6 N.

Cerium carrier, 20 mg/ml.

Hydrochloric acid, 6 N.

Nitric acid, 16 N, 14 N, 4 N, 0.1 N.

Oxalic acid, saturated solution.

Sodium carbonate, 3 N.

Strontium carrier, 88 mg/ml.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

Trichloroacetic acid, 24 per cent (w/v).

Zirconium carrier, 10 mg/ml.

## Procedure

(1) Add 1 ml of strontium carrier to one liter of raw milk and mix thoroughly.

(2) Add 1 liter 24 per cent TCA, stir thoroughly and allow to stand 30 minutes with occasional mixing.

(3) Filter and collect the filtrate, discarding the residue.

(4) Add 90 ml oxalic acid solution and heat to near boiling.

(5) Adjust the pH to 2.5 to 3 with 15 N $NH_4OH$.

(6) Cool and filter through #42 Whatman paper with suction in a Buckner funnel.

(7) Wash precipitate with distilled water.

(8) Dry and ignite precipitate in a muffle furnace for 2 hours at 600°C.

(9) Cool and dissolve the precipitate with 4 N nitric acid and transfer the solution to a 250-ml beaker, washing the ashing vessel thoroughly with 4 N nitric acid.

(10) Evaporate to dryness.

(11) Add 30 ml of 16 N $HNO_3$, heat, and breakup the insoluble nitrates.

(12) Slurry the precipitate and transfer to a 50-ml centrifuge tube, using small portions of concentrated nitric acid to facilitate the transfer.

(13) Cool, centrifuge, and discard the supernate.

(14) Add 30 ml 16 N $HNO_3$, heat in a water bath, stirring frequently and cool in an ice bath.

(15) Centrifuge and discard supernate.

(16) Continue from Procedure (RC-38B-1) step 14.

# SIMPLIFIED DETERMINATION OF STRONTIUM-90 IN MILK

## (RAPID SCREENING TECHNIQUE)

### Principle of Method

Milk proteins are precipitated by trichloroacetic acid (TCA). Calcium is precipitated from the serum at pH 2.5 - 3.0 as the oxalate, carrying strontium and yttrium. Phosphate does not interfere. The oxalate is washed, dried, and ignited to the oxide. The oxide is dissolved in nitric acid and yttrium-90 is extracted with equilibrated TBP. The activity is washed from the organic with 0.1 N $HNO_3$, dried in a stainless steel planchet and counted in a low background beta anti-coincidence counter. Results may be slightly high if the milk contains yttrium-91, zirconium-niobium, or high concentrations of rare-earth radionuclides.

### Reagents

Ammonium hydroxide, 15 N

Ammonium oxalate wash solution, 0.5 per cent.

Nitric acid, 16 N.

Nitric acid - hydrofluoric acid mixture, 14 N $HNO_3$ - 1 N HF.

Oxalic acid solution, saturated.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

Trichloroacetic acid (TCA), 24 per cent (w/v).

### Procedure

(1) Add 1 liter of 24 per cent trichloroacetic acid to 1 liter of milk, let stand 30 minutes, stirring occasionally.

(2) Filter, collecting 1600 ml.

(3) Heat, add 90 ml of hot saturated oxalic acid with stirring.

(4) Adjust the pH to 2.5 to 3.0 with concentrated $NH_4OH$ and allow the sample to digest on a steam bath for one hour.

(5) Decant the supernate and collect the precipitate in a 250-ml centrifuge bottle.

(6) Centrifuge for five minutes and with the aid of vacuum draw off and discard the supernate.

(7) Wash the precipitate 3 times with 0.5 per cent ammonium oxalate solution and discard the supernate.

(8) Slurry the precipitate with 20 ml of 0.5 per cent ammonium oxalate and filter through Whatman #12 fluted filter paper.

(9) Transfer the filter paper and precipitate to a 50-ml evaporating dish. Dry, char, and ignite at 850°C for 3 hours.

(10) Remove the dish from the oven and allow to cool to room temperature.

(11) Place the dish in an ice bath and cautiously add dropwise with stirring 10 ml of 16 $\underline{N}$ $HNO_3$.

(12) Heat the solution on a hot plate until dissolution is completed.

(13) Transfer the solution into a 60-ml separatory funnel and wash the dish with two 10-ml portions of concentrated $HNO_3$.

(14) Add 5 ml of equilibrated TBP and shake thoroughly for 10 minutes.

(15) Transfer the organic layer into a 60-ml separatory funnel and repeat step 14 twice.

(16) Combine the organic layers and add 5 ml 14 $\underline{N}$ $HNO_3$ - 1 $\underline{N}$ HF. Shake thoroughly for 5 minutes and discard the aqueous layer.

(17) Repeat step 16 twice.

(18) Wash 2 more times with 5 ml portions 14 $\underline{N}$ $HNO_3$.

(19) Add 10 ml 0.1 $\underline{N}$ $HNO_3$ to the separatory funnel and shake thoroughly for 10 minutes. Draw off the aqueous layer into a 50-ml beaker.

(20) Repeat step 18 twice.

(21) Transfer the aqueous extracts of steps 19 and 20 into a stainless planchet and evaporate to dryness.

(22) Determine the activity in a low background beta(anti-coincidence) counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(\text{decay factor})(2.22)} = \text{Strontium-90, } \mu\mu c/l \text{ of milk}$$

where:

A = efficiency factor for stainless steel

B = per cent recovery factor (96.0 per cent)

C = aliquot factor = 0.8 (1600/2000)

## Notes

(1) Purity of the yttrium can be determined by decay counting.

(2) The calcium oxalate precipitate carries about 95 per cent of the strontium and 100 per cent of the yttrium. Net loss of yttrium-90 by decay is 1 per cent if the extraction is carried out the day after precipitation of the oxalate.

R.J.V.
11-16-59

# DETERMINATION OF STRONTIUM-90, STRONTIUM-89, BARIUM-140, AND RADIOCESIUM IN MILK

## Principle of Method

Carrier strontium, barium and cesium are added and the milk is ashed by a rapid ashing technique. Radiocesium is separated from barium and strontium by solubilization in nitric acid and by precipitation of barium and strontium as carbonate. Cesium is then recovered by precipitation with ammonium phosphomolybdate followed by purification and final precipitation as the cobaltinitrite, dried and beta counted.

Barium is removed from strontium as the chromate, dried and beta counted.

Strontium is purified by hydroxide scavenging, reprecipitated as the carbonate and allowed to stand for ingrowth of yttrium-90. The precipitate is then dissolved in $HNO_3$ and the yttrium-90 is extracted with tributylphosphate, stripped with 0.1 N $HNO_3$, dried and beta counted.

After the separation of the yttrium, the strontium is again precipitated as the carbonate, dried, weighed and beta counted. Strontium-89 is then determined by difference.

## Reagents

Acetic acid, 18 N, 1.5 N.

Ammonium acetate, 3 N.

Ammonium acetate buffer, pH 5.0.

Ammonium acetate wash solution.

Ammonium hydroxide, 6 N.

Ammonium molybdate, 0.5 N.

Barium carrier, 70 mg/ml.

Barium carrier, 7 mg/ml.

Hydrochloric acid, 6 N.

Iron carrier, 20 mg/ml.

Methyl orange indicator, 0.1 per cent.

Nitric acid, 16 N.

Phosphoric acid, 85 per cent.

Rare earth carrier, mixed.

Sodium carbonate, 3 N.

Sodium cobaltinitrite reagent.

Sodium hydroxide, 6 N.

Sodium chromate, 1 N.

Strontium, barium, and cesium carrier, mixed.

Sodium nitrate, 1 N.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

Procedure

(1) To a liter of whole milk at 20°C add 1 ml of mixed carrier (Sr, Ba, and Cs) and partially ash it according to the rapid ashing technique.

(2) Place the sample in an electric muffle furnace at 450°C for 24 hours to complete the ashing.

(3) Transfer the ash to a 250-ml beaker. Dissolve the residual ash in the ashing crucible with 5-10 ml 6 N HCl by warming, and then transfer the solution to the beaker.

(4) Add 10 ml 6 N HCl to the ash in the beaker, heat on a hot plate to promote solution and continue heating to near dryness.

(5) Add 10 ml 16 N $HNO_3$ and heat to near dryness.

(6) Add 20 ml 16 N $HNO_3$ and heat to near dryness.

(7) Add 40-50 ml 16 N $HNO_3$ and evaporate to about 30 ml (Note 1).

(8) Transfer the slurry into a 50-ml glass centrifuge tube using a little 16 N $HNO_3$ to facilitate transfer. Cool the tube in an ice bath for 15 minutes and then centrifuge. Decant the supernatant and retain it for step 40.

(9) Add 3.0 ml of $H_2O$ and 10 ml 16 N $HNO_3$ to the residue (Note 2). Warm in a water bath, add 20 ml of concentrated $HNO_3$ and continue heating for another 10 minutes with occasional stirring. Cool the solution in an ice bath for 15 minutes and then centrifuge. Transfer the supernatant and combine it with the supernatant retained for step 40.

(10) Wash the remaining residue (Note 3) in the beaker from step 7 into the centrifuge tube with about 10 ml of water. Warm the solution and make it alkaline with an excess of 6 N $NH_4OH$. If no precipitate forms, omit steps 11 and 12 (Note 4).

(11) Centrifuge and decant the supernatant into a 150-ml beaker retaining it for step 14. Add a little water and 10-20 ml 3 N $Na_2CO_3$ to the residue. Digest in a water bath for 30-45 minutes with occasional stirring and then centrifuge. Decant the supernatant, combining it cautiously with the supernatant retained for step 40.

(12) Dissolve the precipitate in 1 to 2 ml 6 N HCl, boil and again test for phosphate with excess of 6 N $NH_4OH$.

(13) If a large precipitate forms, repeat steps 10 and 11. If not (Note 5), combine the supernatant with the supernatant retained for step 14.

(14) Add an excess (5 ml) 3 N $Na_2CO_3$ to the combined solutions (Note 6) with stirring. Centrifuge and discard the supernatant after testing it for completeness of precipitation with $Na_2CO_3$.

(15) Dissolve the precipitate in about 2 ml 6 N HCl, warm to promote solution and to remove any $CO_2$. Neutralize the solution with 6 N $NH_4OH$ using methyl orange as indicator. Dilute to 20-25 ml with water. Add 10 ml of ammonium acetate buffer (pH 5.0). Warm the solution in a water bath, add 1 ml 1 N $Na_2CrO_4$ and digest in the water bath for 10 minutes. Add 1.0 ml 0.1 N $Ba(NO_3)_2$ solution to assure completeness of radiobarium precipitation, and continue digesting for another 10 minutes with occasional stirring. Centrifuge and decant the supernatant, retaining it for step 18 (Note 7).

(16) Wash the $BaCrO_4$ precipitate with 5 ml $H_2O$, warm, centrifuge and decant the washing, combining it with the supernatant from step 15.

(17) Transfer the $BaCrO_4$ precipitate quantitatively to an aluminum planchet, dry, cool and determine beta activity (Note 8).

(18) Make the combined supernatants alkaline with excess 6 N $NH_4OH$ and precipitate strontium with 3-5 ml 3 N $Na_2CO_3$. Centrifuge and discard the supernatant after testing it for completeness of precipitation with $Na_2CO_3$.

(19) Wash the precipitate once with water containing a drop of $Na_2CO_3$. Centrifuge and discard the washings.

(20) Dissolve the precipitate in about 2-3 ml 6 N HCl, dilute to 15 ml with water. Add 1-2 drops of iron solution, heat it in a water bath and then make the solution alkaline with excess 6 N $NH_4OH$. Digest for 5 minutes. Centrifuge and retain supernate. Wash precipitate by dissolving and reprecipitating in 10 ml distilled water. Combine washing with supernate. Discard precipitate.

(21) Add to the supernate 1-2 drops of cerium and zirconium carrier solution, dissolve any precipitate that forms with 6 N HCl. Warm the solution and make it alkaline with $NH_4OH$. Centrifuge and retain supernate. Wash precipitate by dissolving and reprecipitating in 10 ml of distilled water. Combine washing with supernate. Discard precipitate. Record the time (Note 9).

(22) Add to the supernate about 5 ml 3 N $Na_2CO_3$ to precipitate the strontium. Centrifuge and discard the supernatant after testing it for completeness of precipitation. Wash the precipitate with water containing a drop of $Na_2CO_3$ and discard the washing (Note 10). Set aside the precipitate for two weeks for ingrowth of yttrium-90 (Note 11).

(23) Dissolve the precipitate in 2 ml 4 N $HNO_3$.

(24) Cool in an ice bath and add with stirring 10 ml of fuming nitric acid (Note 12).

(25) Centrifuge and transfer the supernate into a 60-ml separatory funnel.

(26) Dissolve the residue in 2 ml 4 N $HNO_3$ and repeat steps 24 and 25.

(27) Save precipitate for step 38 (Note 13).

(28) Add 6 ml 4 N $HNO_3$ to the separatory funnel to bring the aqueous phase to 30 ml.

(29) Add 5.0 ml of equilibrated TBP and shake thoroughly for 10 minutes.

(30) Transfer the organic layer to another separatory funnel and repeat step 29 twice.

(31) Wash the combined organic phases three times with 5 ml portions 14 N $HNO_3$. Discard the washings.

(32) Back extract the combined organic phases with 10 ml 0.1 N $HNO_3$ for 10 minutes.

(33) Transfer the aqueous phase to a 50-ml beaker and reduce its volume on a hot plate.

(34) Repeat steps 32 and 33 twice, combining the aqueous phases in the 50-ml beaker.

(35) Evaporate the solution to about 10 ml and transfer it into a stainless steel planchet and evaporate to dryness.

(36) Wash beaker several times with small volumes (2 ml) 0.1 N $HNO_3$.

(37) Determine the activity in a low background beta (anti-coincidence) counter.

(38) Dissolve precipitate from step 27 with 15 ml water, heat and add $NH_4OH$ to make basic and precipitate strontium with 5 ml 3 N $Na_2CO_3$. Stir, centrifuge and discard supernate.

(39) Wash the precipitate, transfer it to a tared aluminum planchet, dry, cool, weigh, and determine the beta activity of the total strontium.

(40) Evaporate the supernatant from steps 8, 9, and 11 almost to dryness to get rid of excess nitric acid.

(41) Dissolve the residue in about 100 ml of 10 per cent $HNO_3$, add 0.5 ml of phosphoric acid.

(42) Warm the solution to 50°C, add 10 ml of ammonium molybdate solution, stir the solution mechanically for at least 1/2 hour.

(43) Remove the cesium ammonium phosphomolybdate precipitate by filtration through a sintered glass (fine grade) funnel. Wash the precipitate three times by decantation, each time with 25 ml of 1 N sodium nitrate solution.

(44) Dissolve the precipitate in the beaker and the filtering crucible with 5 ml of 6 N NaOH. Collect the solution in a

50-ml glass centrifuge tube. Wash the beaker and the crucible several times with a small quantity of distilled water and collect the washings in the same centrifuge tube.

(45) Add 2 grams of $NaNO_2$ and 5 ml of glacial acetic acid to the solution and heat it in a boiling water bath for at least 10 minutes (Note 14).

(46) Cool the solution in an ice bath, add 3 ml of sodium cobaltinitrite solution and allow it to stand in the ice bath for 10 minutes with occasional stirring.

(47) Remove the precipitate by centrifugation, discard the supernatant after testing it for completeness of precipitation.

(48) Wash the precipitate with 10 ml of distilled water at 10°C and discard the washings.

(49) Transfer the precipitate to a tared aluminum planchet, dry, cool, and weigh.

(50) Determine the beta activity.

Notes

(1) It will be helpful to make a rough calibration of the beaker beforehand.

(2) The residue is largely strontium and barium nitrates.

(3) The residue may still contain a little precipitate which was not previously transferred.

(4) A precipitate at this point indicates unremoved phosphate.

(5) A small precipitate of silica may be observed and is discarded.

(6) These solutions contain strontium and barium.

(7) Barium is separated from strontium in steps 15 through 17.

(8) For beta counting (Ba = La) optimum waiting time is 94 hours and for gamma counting of lanthanum-140, the optimum waiting time is 136 hours.

(9) This is the time from which the ingrowth of yttrium-90 is calculated.

(10) Total strontium activity may be determined at this time.

(11) The strontium is left in a solid state so that yttrium-90 formed may be trapped in the crystal minimizing adsorption losses.

(12) This is the end of yttrium-90 ingrowth and the beginning of yttrium-90 decay.

(13) This total strontium analysis should be performed immediately to minimize yttrium-90 ingrowth contribution.

(14) Sodium nitrite is used to destroy ammonium ions.

(15) Chemical yield is determined gravimetrically or by flame photometry.

Calculation

a. Strontium-90

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(F)(2.22)} = \text{Strontium-90, } \mu\mu c/l$$

where:

A = efficiency factor on stainless steel

B = per cent extraction factor (0.98)

C = per cent ingrowth factor (0.97)

D = chemical yield (Note 15)

E = volume, liters

F = decay factor; calculated from the following formula:

$$A = A_0 e^{-\lambda t}$$

where:

A = activity remaining after a time interval, t

$A_o$ = activity of sample at original time (step 24)

$\lambda = \frac{.693}{\text{half life of yttrium (64.2 hrs)}}$

t = time (hours) from separation to counting

b. Strontium-89

$$\frac{\text{Net cpm} - (A)}{(B)(C)(D)(E)(2.22)} = \text{Strontium-89, } \mu\mu c/l$$

where:

A = net cpm due to strontium-90 equivalent to 1PC
B = chemical yield (Note 15)
C = self absorption factor for strontium-89
D = efficiency factor
E = volume, liters

c. Barium-140

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(2.22)} = \text{Barium-140, } \mu\mu c/l$$

where:

A = efficiency factor
B = self absorption factor
C = chemical yield (Note 15)
D = volume, liters

d. Radiocesium

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)(1.08)} = \text{Radiocesium, } \mu\mu c/l$$

where:

A = efficiency factor
B = self absorption factor
C = volume, liters

References

1. Murthy, G. K., and Campbell, J. E., "A Method for the Rapid Ashing of Milk for Radionuclide Analysis," Journal of Dairy Science, Vol. XLII, No. 8, 1288-1293, August (1959).

2. Goldin, A. S., Murthy, G. K., "A Method for the Determination of Total Strontium, Strontium-90, Barium-140, Cesium-137, and Iodine-131," Radiological Health Program, Robert A. Taft Sanitary Engineering Center, U. S. Public Health Service, Cincinnati 26, Ohio.

# DETERMINATION OF STRONTIUM-90 AND RADIUM-226 IN BONE

## Principle of Method

Strontium, barium, and lead are added as carriers for strontium-90 and radium-226. Calcium is precipitated as the oxalate, eliminating phosphate. Strontium and radium are separated from most of the calcium by precipitation as the nitrate. Radium is separated by precipitation as the chromate, converted to sulfate and purified by solution and re-precipitation in an EDTA cycle. It is then determined by alpha counting.

The solution, remaining after removal of radium with barium and lead, is purified by hydroxide scavenging and the strontium is precipitated as the carbonate and allowed to stand for ingrowth of yttrium-90. The activity (yttrium-90) is extracted with equilibrated TBP, washed from the organic with 0.1 N $HNO_3$, dried in a stainless steel planchet and counted in a low background beta anti-coincidence counter.

## Reagents

Acetic acid, 18 N.

Acetone, reagent grade.

Ammonium acetate buffer, pH 5.0.

Ammonium hydroxide, 15 N, 6 N.

Ammonium oxalate, saturated.

Ammonium oxalate wash solution, 0.5 per cent.

Barium carrier, 20 mg/ml.

Ethylenediaminetetraacetic acid, (EDTA) M/4.

Hydrochloric acid, 6 N.

Lead carrier, 100 mg/ml.

Methyl orange indicator, 0.1 per cent.

Nitric acid, fuming, 16 N, 14 N, 8 N, 0.1 N.

Potassium chromate, 1.0 N.

Sulfuric acid, 18 N.

Sodium carbonate, 3 N.

Strontium carrier, 88 mg/ml.

Rare earth carrier, mixed.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

Preparation of Sample

The bone samples are received in heat-sealed polyethylene bags. They have been treated with paraformaldehyde as a preservative during shipment.

The bone sample is removed from the bag, washed with distilled water and placed in a beaker. The beaker is covered with aluminum foil and autoclaved for one hour, after which the soft tissue is easily separated from the bone. The bone is dried in an oven at 105°C.for four hours and cooled to room temperature. The sample is then weighed and ashed in a muffle furnace, the temperature being slowly raised from 100°C to 750°C.

Procedure

a. Strontium-90

(1) Transfer a 35-gram sample of bone ash into a one liter beaker (Note 1).

(2) Slowly add with stirring 100 ml of concentrated $HNO_3$.

(3) Heat on a hot plate until dissolution is complete.

(4) Add 1.0 ml of strontium and barium carrier and 2.0 ml of lead carrier.

(5) Add 250 ml of distilled water and heat to near boiling.

(6) Add 450 ml of hot saturated ammonium oxalate with stirring.

(7) With the aid of a pH meter, adjust the pH to 3.0 with concentrated $NH_4OH$ and allow the sample to digest on a steam bath for one hour.

(8) Allow the precipitate to settle and draw off the supernate by vacuum into a suction flask. Save the supernate for the determination of radium. (Sub procedure b.)

(9) Transfer the precipitate into four 250 ml centrifuge bottles.

(10) Centrifuge for five minutes and with the aid of vacuum draw off and combine the supernates with the supernate from step 8.

(11) Wash the precipitates once with 20 to 30 ml 0.5 per cent ammonium oxalate solution and combine the supernates with the supernate from step 8.

(12) Slurry the precipitates with 50 ml 0.5 per cent ammonium oxalate solution and filter through Whatman #12 fluted filter paper. Combine the filtrates with the supernate retained from step 8.

(13) Transfer the filter papers and precipitates to tared 50 ml evaporating dishes. Dry, char, and ignite at 750°C for three hours.

(14) Remove the dishes from the oven and allow them to cool to room temperature.

(15) Weigh the dishes and CaO on an analytical balance and record the CaO weight.

(16) Place the dishes in an ice bath and slowly with stirring add enough 8 N nitric acid to dissolve the oxide (Note 2). Transfer the solution into a 250 ml centrifuge bottle.

(17) Add five times the volume of fuming $HNO_3$ as was required of 8 N nitric acid to dissolve the calcium oxide (Note 3).

(18) Cool the nitrate precipitate in an ice bath for several minutes and centrifuge, collecting the precipitate in 250 ml centrifuge bottles.

(19) Discard the supernatant. Invert the centrifuge bottle on absorbent paper and allow the excess nitric acid to drain completely. Cool the precipitate in an ice bath and cautiously add with stirring 20 ml of cold anhydrous acetone. Centrifuge and discard supernatant.

(20) Dissolve the nitrate precipitate with distilled water and make the solution alkaline with concentrated $NH_4OH$.

(21) Precipitate carbonates with an excess of sodium carbonate and collect the precipitate in a 250 ml centrifuge bottle. Discard the supernatant after testing for completeness of precipitation.

(22) Dissolve the precipitate with 6 N HCl.

(23) Add a few drops of methyl orange indicator and add 6 N $NH_4OH$ dropwise to a methyl orange end point.

(24) Slowly and with stirring add 5 ml acetate buffer solution. Digest for about 10 minutes.

(25) Add 1.0 ml of $K_2CrO_4$ and warm in a water bath for several minutes.

(26) Centrifuge and transfer the supernatant into a clean 250 ml centrifuge bottle.

(27) Dissolve the lead and barium chromate precipitate with 6 N HCl and combine with the supernatant from step 8, being retained for radium analysis.

(28) Make the solution from step 26 alkaline with concentrated ammonium hydroxide and add 1.0 ml of mixed rare earth carrier. Digest in a hot water bath for about 10 minutes (Note 4).

(29) Centrifuge and discard the precipitate.

(30) Repeat the rare earth scavenge in step 28, centrifuge and discard the precipitate.

(31) Add sufficient $Na_2CO_3$ for complete precipitation of strontium carbonate (Note 5).

(32) Store this precipitate for two weeks.

(33) Dissolve the precipitate with concentrated $HNO_3$ and transfer the solution into a 250 ml separatory funnel. Adjust the volume to 120 ml with concentrated $HNO_3$.

(34) Add 20 ml of equilibrated TBP and shake thoroughly for 10 minutes (Note 6).

(35) Transfer the organic layer into a 125 ml separatory funnel and repeat step 34 twice.

(36) Combine the organic layers and add 20 ml 14 N $HNO_3$. Shake thoroughly for 5 minutes and discard the aqueous layer.

(37) Repeat step 36 twice.

(38) Add 40 ml of distilled water to the separatory funnel and shake thoroughly for 10 minutes. Draw off the aqueous layer into a 250 ml beaker.

(39) Add 40 ml 0.1 N nitric acid to the separatory funnel and shake thoroughly for 10 minutes. Combine the aqueous layer in the 250 ml beaker.

(40) Repeat step 39 twice.

(41) Transfer the solution into a stainless steel planchet and evaporate to dryness.

(42) Determine the activity in a low background beta (anti-coincidence) counter.

Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(F)(2.22)} = \text{Strontium-90, } \mu\mu c/g \text{ of ash}$$

where:

A = efficiency factor for stainless steel in low background beta counter

B = per cent extraction factor (0.98)

C = per cent ingrowth factor (0.97)

D = chemical yield (Note 7)

E = sample weight, grams

F = decay factor; calculated from the following formula:

$$A = A_o e^{-\lambda t}$$

where:

$A$ = activity remaining after a time interval, t

$A_o$ = activity of sample at some original time

$\lambda = \frac{0.693}{\text{half-life of yttrium (64.2 hrs)}}$

t = time (hours) from separation to counting

Notes

(1) Where a smaller sample size is used correspondingly smaller volumes of reagents can be used throughout the procedure.

(2) The calcium oxide may be wetted with a little distilled water to inhibit the initial reaction.

(3) The normality of the solution at this point should be at least 17 N.

(4) The rare earth scavenge is carried out to insure the separation of strontium and yttrium.

(5) This is the start of ingrowth time for Y-90.

(6) This is the end of ingrowth time for Y-90; start of decay time for Y-90.

(7) Chemical yield is determined by flame photometry.

b. Radium-226

(1) Heat the supernatant saved from the strontium-90 determination to boiling and add 3 to 5 ml 18 N sulfuric acid (Note 1).

(2) Allow the precipitate to settle and siphon off the supernatant and discard it.

(3) Collect the precipitate in a 50 ml centrifuge tube.

(4) Wash the precipitate twice with 10 ml portions 15 N $HNO_3$, and discard the washings.

(5) Dissolve the precipitate in 10 ml distilled water, 10 ml EDTA, and 3 ml 6 N ammonium hydroxide. Warm and add dropwise to the warm solution 2 ml 18 N acetic acid (Notes 2 and 3).

(6) Digest 5 - 10 minutes, centrifuge, and discard the supernate.

(7) Wash the reprecipitated barium sulfate with water and transfer it into a centrifuge tube adapted for centrifuging precipitates onto planchets (Note 4).

(8) Allow the precipitate to settle 5 - 10 minutes, centrifuge, dry, flame, weigh, and determine the alpha activity in an internal proportional counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(2.22)} = \text{Radium-226 } \mu\text{pc/g of ash}$$

where:

A = ingrowth factor (Note 5)

B = efficiency factor for alpha counting

C = self absorption factor

D = chemical yield

E = sample weight, grams

## Notes

(1) If a residue remains after boiling, add $HNO_3$ to dissolve. Add the sulfuric acid as prescribed and then with the aid of a pH meter adjust the pH to 1.0 with 15 N $NH_4OH$.

(2) This quantity of acetic acid is in approximately 2:1 excess over the $NH_4OH$, giving a pH of about 4.5. This is sufficient to destroy the Ba-EDTA complex, but not the Pb-EDTA.

(3) From this time on, Rn (and daughters) grow into the $BaSO_4$ precipitate.

(4) Atomlab-Eckstein centrifuge tube. If not available, the precipitate can be transferred in any suitable manner.

(5) Can be calculated from data of Kirby, in AEC Report MLM-859, (see RC-88A-1).

## References

1. Goldin, A. S., Velten, R. J., and Frishkorn, G. W., "Determination of Radioactive Strontium", Anal. Chem. 31, 1490 (1959).

2. "Analysis of Water for Dissolved Radium-226," Goldin, A. S., In Charge Radiochemical Methods, Radiological Health Program, Robert A. Taft Sanitary Engineering Center, U. S. Public Health Service, Cincinnati 26, Ohio.

# DETERMINATION OF STRONTIUM-90 IN AQUATIC BIOTA, VEGETATION, SILT AND SOIL

## Principle of Method

The sample is converted to ash, usually in a muffle furnace. For some samples, however, wet ashing with nitric acid followed by an equal volume mixture of nitric and perchloric acids may be preferred. A suitable aliquot is taken, dissolved in hydrochloric acid, and evaporated to near dryness. The chlorides are converted to nitrates by evaporation with nitric acid and strontium nitrate is precipitated from concentrated nitric acid. After a second precipitation from nitric acid, the precipitate is dissolved in water and made alkaline with ammonia to test for completeness of phosphate removal. Any precipitate which forms (indicative of unremoved phosphate) is digested with sodium carbonate to convert the phosphate to carbonate. This is dissolved in acid, warmed to drive off carbon dioxide and again made alkaline with ammonium hydroxide. If a precipitate forms, it is discarded. Sodium carbonate is added to the combined strontium-containing supernatants to precipitate the strontium as carbonate. This precipitate is collected and purified by barium chromate precipitation and hydroxide scavenging. In all cases with biological samples, treatment with nitric acid is necessary to prevent phosphate interference.

## Reagents

Ammonium acetate buffer, pH 5.0.

Ammonium acetate wash solution.

Ammonium hydroxide, 15 N, 6 N.

Barium carrier, 70 mg/ml.

Cerium carrier, 20 mg/ml.

Hydrochloric acid, 12 N, 6 N, 1 N.

Iron carrier, 50 mg/ml.

Methyl orange indicator, 0.5 per cent.

Nitric acid, fuming, 16 N, 14 N, 4 N, 0.1 N.

Perchloric acid, 70-72 per cent.

Potassium chromate, 1.0 N.

Sodium carbonate, reagent grade.

Sodium carbonate, 3 N.

Sodium hydroxide, reagent grade.

Sodium hydroxide, 19 N, 6 N.

Strontium carrier, 88 mg/ml.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

Zirconium carrier, 10 mg/ml.

## I. AQUATIC BIOTA AND VEGETATION

### Procedure

(1) Dry the sample at 105°C and then ash in a muffle furnace at 600°C (Note 1).

(2) Transfer a 10-gram aliquot sample of ash into a 250-ml beaker.

(3) Add 2.0 ml of strontium carrier and add 20 ml 6 N HCl. Heat and evaporate to near dryness.

(4) Add 10 ml of 16 N $HNO_3$ and heat to near dryness.

(5) Add 20 ml 16 N $HNO_3$ and heat to near dryness.

(6) Add 40-50 ml 16 N $HNO_3$ evaporate to 25-30 ml.

(7) Transfer to a 50-ml centrifuge tube (transfer of solid need not be quantitative); cool 5-10 minutes in an ice bath, centrifuge decant, and discard the supernatant.

(8) Transfer the solids remaining in the beaker (step 7) to the centrifuge tube with 16 N $HNO_3$, add additional 16 N $HNO_3$ to a volume of about 30 ml.

(9) Warm in a water bath 10-15 minutes with occasional stirring, cool 5-10 minutes in an ice bath, centrifuge, decant the supernatant, and discard.

(10) Take up the solid with about 10 ml of water, warm and make alkaline with 6 N $NH_4OH$. If no precipitate forms, omit steps 11-16.

(11) Centrifuge, decant, and retain the supernatant.

(12) Wash the residue from the centrifuge tube into a beaker with 10-20 ml 3 N $Na_2CO_3$. Boil gently about 30 minutes, stirring from time to time.

(13) Return the mixture from step 12 to the centrifuge tube. Water may be used in the transfer as necessary. Centrifuge and discard the supernatant.

(14) Dissolve the precipitate from step 13 in a few ml of 6 N HCl and about 10 ml of $H_2O$, heat, add 6 N $NH_4OH$ in excess.

(15) If a large precipitate forms, repeat steps 11-14.

(16) If a small (or no) precipitate forms, discard it and combine the supernatant with that obtained in step 11.

(17) To the liquid from step 10, or the combined supernatants of steps 11 and 16, add enough 3 N $Na_2CO_3$ solution to precipitate all of the alkaline earths.

(18) Collect the precipitate in a centrifuge tube and discard the supernatant.

(19) Dry the precipitate thoroughly in an oven at 100°C.

(20) Remove, cool, and add cautiously 30 ml 16 N $HNO_3$ and warm in a water bath about 10 minutes, stirring occasionally.

(21) Cool in an ice bath about 5 minutes, stirring occasionally.

(22) Centrifuge and discard the supernatant.

(23) Repeat steps 20-22.

(24) Take up the $Sr(NO_3)_2$ precipitate in 20 ml of water and transfer it into a 50-ml centrifuge tube.

(25) Make alkaline with 6 N $NH_4OH$ and reprecipitate the strontium with 5 ml of 3 N $Na_2CO_3$.

(26) Centrifuge, discard the supernatant. Wash the precipitate with water containing a drop or so of 3 N $Na_2CO_3$ solution and discard the washings.

(27) Continue with steps 13 through 34 of RC-38B-1.

Notes

(1) Some samples are preferably wet ashed as follows:

a. Add cautiously to the sample (10 grams or less in a 250-ml beaker) 2.0 ml of 1 N $Sr(NO_3)_2$ and 10-20 ml of $HNO_3$. Evaporate dry or nearly dry. Caution is needed here, since some samples will tend to foam, especially in the early stages.

b. Add a mixture of 10 ml 16 N $HNO_3$ and 10 ml of 70 per cent $HClO_4$. Heat to dense fumes of $HClO_4$ and reduce the volume to about 5 ml.

c. Repeat previous step as often as it appears useful. Samples should become water-white unless iron or other colored non-organic materials are present.

d. Cool, add 20-25 ml of $H_2O$, make alkaline with 6 N $NH_4OH$, add enough 3 N $Na_2CO_3$ solution to complete precipitation, centrifuge, and discard supernatant.

e. Transfer the precipitate into a 250-ml beaker and proceed with step 4.

## II. SILT AND SOIL

Procedure

(1) Ash the sample in a muffle furnace at 600°C.

(2) Take an aliquot of 10 grams of ash (Note 1).

(3) Add 2.0 ml of 1 N $Sr(NO_3)_2$, mix uniformly, and dry at 10.5°C.

(4) Mix the sample thoroughly with 50 grams of solid NaOH and fuse in a nickel crucible.

(5) Add to the melt 5 grams of solid $Na_2CO_3$.

(6) Continue heating until the melt is clear.

(7) Take up the cooled fusion mixture in hot water (Note 2).

(8) Centrifuge and discard the supernatant.

(9) Dissolve the residual solid in 6 N HCl. Warm to drive off $CO_2$.

(10) Make the solution alkaline with excess 6 N $NH_4OH$.

(11) Separate the precipitate by filtration or centrifugation. Retain the liquid phase.

(12) Dissolve the precipitate from step 11 in 6 N HCl, and re-precipitate with excess 6 N $NH_4OH$.

(13) Repeat step 11, combining this liquid phase with the one obtained in step 11.

(14) Repeat steps 12 and 13. Discard the precipitate (Note 3).

(15) Add enough 3 N $Na_2CO_3$ solution to the combined liquid phases (from steps 11, 13, and 14) to precipitate the alkaline earths completely, as shown by failure to obtain more precipitate on addition of more 3 N $Na_2CO_3$.

(16) Collect the precipitate in a centrifuge tube, discard the supernatant.

(17) Follow steps 18 through 27 of the procedure outlined in Part I. Aquatic Biota and Vegetation.

Notes

(1) Larger aliquots may be taken, up to 50 g, provided appropriate changes are made in quantities of reagents. Quantities of reagents may also have to be changed for silts or soils of unusual types.

(2) Difficulty in disintegrating the fusion mixture may be avoided by the following technique:

a. Carefully pour the molten material into a clean nickel crucible and then cool the outside of this second crucible to break the melt away from the wall.

b. Transfer the melt to a beaker containing about 250 ml of $H_2O$ and boil to complete the disintegration. Any fused material remaining in either crucible is readily washed out with hot water and added to the beaker.

(3) Two cycles of solution and reprecipitation are given to wash the hydroxide precipitate. More cycles may be added as desired in case of an extremely bulky precipitate.

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# DETERMINATION OF STRONTIUM-90, STRONTIUM-89, GROSS BETA (BASIC SULFIDE) AND RADIOCESIUM IN RIVER WATER

## DETERMINATION OF STRONTIUM-90

### Principle of Method

Carrier strontium is added and precipitated as the carbonate. The precipitate is dissolved in nitric acid and purified by hydroxide scavenging. The strontium is then precipitated from the supernatant solution with carbonate and allowed to stand for ingrowth of yttrium-90. The precipitate is then dissolved in nitric acid and the yttrium-90 is extracted with equilibrated TBP, washed from the organic with 0.1 N $HNO_3$, dried in a stainless steel planchet and counted in an internal proportional counter.

### Reagents

Ammonium hydroxide, 15 N, 6 N.

Mixed carrier solution, Sr, 22 mg; Cs, 3.25 mg; Fe, 1.25 mg; Zr, 1.25 mg; Zn, 1.25 mg; Ce, 1.25 mg; and Co, 1.25 mg per ml.

Nitric acid, fuming, 4 N, 0.1 N.

Sodium carbonate, 3 N.

Sodium hydroxide, 6 N.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$.

### Procedure

(1) Transfer a one liter composite sample* into a 1500-ml beaker and add 4.0 ml mixed carrier solution.

(2) Add 10 ml 6 N NaOH and heat to near boiling.

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* If the activity is known to be low, a 12-liter sample can be taken and reduced by evaporation to a two liter volume. One liter aliquots can then be analyzed in duplicate.

(3) Add 10 ml 3 N $Na_2CO_3$ with stirring and allow the precipitate to settle until the supernate is clear.

(4) Siphon off the bulk of the supernate (save for basic sulfide and cesium determinations), and collect the precipitate in a 50-ml cone shaped centrifuge tube.

(5) Dry the precipitate and original beaker.

(6) Wash the original beaker with 20 ml 15 N $HNO_3$ in two 10 ml portions.

(7) Carefully add the washings to the dried precipitate. (Add a little at a time until all of the carbonate has reacted).

(8) Heat in a water bath until the supernate is clear and then cool in an ice bath. Centrifuge and add the supernate to the original solution from step 4.

(9) To the precipitate, add 30 ml 15 N $HNO_3$ and repeat step 8.

(10) Dissolve the precipitate in water.

(11) Make the solution alkaline with 15 N $NH_4OH$ to precipitate the insoluble hydroxides. Stir and centrifuge, transferring the supernatant into a clean 50-ml centrifuge tube. Discard the residue.

(12) Add sufficient $Na_2CO_3$ for complete precipitation of strontium carbonate (Note 17).

(13) Store this precipitate for two weeks.

(14) Dissolve the precipitate in 2 ml 4 N $HNO_3$.

(15) Cool in an ice bath and add with stirring 10 ml of fuming nitric acid.

(16) Centrifuge and transfer the supernate into a separatory funnel.

(17) Dissolve the residue in 2 ml 4 N $HNO_3$ and repeat steps 15 and 16. Save the residue for the determination of strontium-89 (Note 2).

(18) Add 6 ml 4 N $HNO_3$ to the separatory funnel to bring the aqueous phase to 30 ml.

(19) Add 5.0 ml of equilibrated TBP and shake thoroughly for 10 minutes.

(20) Transfer the organic layer into another 60-ml separatory funnel and repeat step 19 twice.

(21) Wash combined organic phases three times with 5-ml aliquots 14 N $HNO_3$.

(22) Back extract the combined organic phases with 10 ml 0.1 N $HNO_3$ for 10 minutes.

(23) Transfer the aqueous phase to a 50-ml beaker and reduce its volume on a hot plate.

(24) Repeat steps 22 and 23 twice, combining the aqueous phases in the 50-ml beaker.

(25) Evaporate the solution to about 10 ml and transfer it into a stainless steel planchet and evaporate to dryness.

(26) Wash beaker several times with small aliquots 0.1 N $HNO_3$.

(27) Determine the beta activity in an internal proportional counter.

Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(D)(E)(F)(2.22)} = \text{Strontium-90, } \mu\mu c/l$$

where:

A = efficiency factor on stainless steel

B = per cent extraction factor (0.98)

C = per cent ingrowth factor (0.97)

D = chemical yield (Note 3)

E = sample volume, liters

F = decay factor; calculated from the following formula:

$$A = A_o e^{-\lambda t}$$

where:

$A$ = activity remaining after a time internal, t

$A_o$ = activity of sample at some original time

$\lambda = \frac{0.693}{\text{half life of yttrium (64.2 hrs.)}}$

t = time (hours) from separation to counting

Notes

(1) This is the start of ingrowth time for yttrium-90.

(2) This is the end of ingrowth time for yttrium-90; start of decay time of yttrium-90.

(3) Chemical yield is determined gravimetrically or by flame photometry.

## DETERMINATION OF STRONTIUM-89

### Principle of Method

After the separation of yttrium from the strontium by nitrate precipitation, the strontium nitrate is immediately reprecipitated as the carbonate and the activity of the total strontium is determined. Strontium-89 is then determined by difference.

### Reagents

Sodium carbonate, 3 N.

### Procedure

(1) To the residue from step 17 (determination of strontium-90) add 15 ml of distilled water and dissolve the residue. Make the solution alkaline with $NH_4OH$.

(2) Slowly add sufficient $Na_2CO_3$ for complete precipitation of strontium carbonate.

(3) Centrifuge, check for completeness of precipitation, decant and discard the supernate.

(4) Slurry, with the aid of a wash bottle, and transfer the precipitate into an aluminum planchet.

(5) Dry the precipitate in an oven at 100°C.

(6) Cool, and place the planchet in an internal proportional counter and determine the beta activity.

Calculation

$$\frac{\text{Net cpm} - (A)}{(B)(C)(D)(E)(2.22)} = \text{strontium-89, } \mu\mu c/l$$

where:

A = net cpm due to strontium-90 (corrected for self absorption)

B = chemical yield

C = self absorption factor for strontium-89

D = efficiency factor for aluminum (0.64)

E = sample volume, liters.

DETERMINATION OF GROSS BETA ACTIVITY (BASIC SULFIDE)

Principle of Method

The determination of gross radioactivity is based on the precipitation of many cations as sulfides from ammoniacal solution. Carrier iron (III), cobalt (II), and zinc (II) are precipitated. This precipitate, containing the radioactivity other than that attributed to cesium and the alkaline earth fraction, is dried, ignited, and counted.

Reagents

Ammonium hydroxide, 6 N.

Hydrogen sulfide, commercial grade.

Procedure

(1) Make the supernate, obtained from steps 4, 8, and 9 (determination of strontium-90) alkaline with 15 N $NH_4OH$, heat to near boiling and add $H_2S$ until precipitation is complete.

(2) Filter off the precipitate (Whatman 41), wash with hot water and save the filtrate for the subsequent determination of cesium.

(3) Char and ignite the precipitate and paper at 650°C for one hour.

(4) Cool and transfer the residue with the aid of a wash bottle into a clean tared aluminum planchet.

(5) Dry and weigh the precipitate.

(6) Place the planchet in an internal proportional counter and determine the beta activity.

Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)} = \text{activity, } \mu\mu c/l$$

where:

A = efficiency factor for aluminum (0.624)

B = self absorption correction factor

C = volume, liters

DETERMINATION OF RADIOCESIUM

Principle of Method

After removal of the alkaline earths and sulfides, the cesium is recovered separately by precipitation with ammonium phosphomolybdate, followed by purification and final precipitation as the cobaltinitrite. The precipitate of cesium cobaltinitrite is counted for beta activity in an internal proportional counter.

Reagents

Acetic acid, 18 N.

Ammonium hydroxide, 15 N.

Ammonium molybdate, 0.08 N.

Nitric acid, 16 N.

Phosphoric acid, 85 per cent.

Sodium nitrite, reagent grade.

Sodium cobaltinitrite reagent.

Procedure

(1) To the filtrate from the basic sulfide determination (step 2), slowly add 50 ml 16 N·$HNO_3$.

(2) Add 0.5 ml $H_3PO_4$ and heat the solution at 50°C to expel any $H_2S$.

(3) Add 40 ml 10 per cent ammonium molybdate and stir vigorously for about thirty minutes. (Use a magnetic stirrer.)

(4) Allow the precipitate of cesium ammonium phosphomolybdate to settle.

(5) Siphon or decant the bulk of the supernatant and discard it.

(6) Collect the precipitate on a fine fritted disc filter funnel and discard the filtrate.

(7) Place a 50-ml centrifuge tube under the funnel and add 5 to 10 ml 6 N NaOH directly into the fritted disc funnel several minutes to dissolve the precipitate.

(8) With the aid of suction draw the solution into the centrifuge tube.

(9) Wash the filter bed with a little water and suck the washings into the centrifuge tube.

(10) Add 5 ml 18 N acetic acid, with stirring, and add 2 grams of sodium nitrite.

(11) Digest in a water bath for about 10 minutes.

(12) Cool in an ice bath and add 2 ml of $Na_3Co(NO_2)_6$ reagent to precipitate cesium cobaltinitrite.

(13) Wash precipitate with 10 ml $H_2O$ at 10°C.

(14) Transfer the precipitate to an aluminum planchet. Dry at 100°C and count the activity in an internal proportional counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)(1.08)} = \text{radiocesium, } \mu\mu c/l$$

where:

A = self absorption factor

B = efficiency factor

C = volume, liters

## References

1. Kahn, B., Smith, D. K., and Straub, C. P., Anal. Chem. 29, 1210, August (1957).

2. Goldin, A. S., Velten, R. J., and Frishkorn, G. W., "Determination of Radioactive Strontium," Anal. Chem. 31, 1940 (1959).

# DETERMINATION OF IODINE-131 IN WATER

## Principle of Method

Carrier sodium iodide is oxidized by sodium hypochlorite in basic solution to iodate and periodate. It is then reduced to iodide by sodium bisulfite in acid solution. This procedure assures interchange of carrier and radioiodine. Sodium nitrite is then added to oxidize to elemental iodine, which is extracted into carbon tetrachloride. This serves as both a purification and concentration step. The iodine is further purified and concentrated by back-extraction into sodium bisulfite solution which is finally gamma-counted at the I-131 photo peak.

## Reagents

Carbon tetrachloride, reagent grade.

Hydrochloric acid, 6 N.

Iodine carrier, 13 mg/ml.

Sodium bisulfite, 1 N.

Sodium hydroxide, 6 N.

Sodium hypochlorite, 5.25 per cent. (Note 1.)

Sodium nitrite, 1 N.

## Procedure

(1) Transfer a 500-ml aliquot of sample into a one liter beaker.

(2) Add 1.0 ml of iodine carrier, 5 ml 6 N NaOH, 1 ml NaClO (Note 1) and heat to near boiling.

(3) Transfer the solution to a separatory funnel and add 10 ml 6 N HCl (Note 2).

(4) Add dropwise sufficient 1 N $NaHSO_3$ to reduce all of the iodate to iodide.

(5) Add 5 ml $CCl_4$ and 3 ml 1 N $NaNO_2$. Shake thoroughly and draw off the organic layer into a 60-ml separatory funnel containing 2 ml 1 N $NaHSO_3$ solution.

(6) Repeat step 5 using 1 ml of 1 N $NaNO_2$.

(7) Repeat step 6 twice and discard the aqueous layer (Note 3).

(8) Shake the separatory funnel thoroughly until the $CCl_4$ layer is decolorized, allow the phases to separate, and transfer the aqueous layer to a counting tube (Note 4).

(9) Add 2 ml 1 N $NaHSO_3$ to the separatory funnel and repeat step 8.

(10) Add 1 ml $H_2O$ to the separatory funnel and repeat step 8.

(11) Count the activity contained in the combined aqueous extracts in a well-type gamma scintillation counter.

## Calculation

$$\frac{\text{Net cpm}}{(A)(B)(C)(2.22)} = \text{Iodine-131, } \mu\mu c/l$$

where:

A = counting efficiency
B = chemical yield
C = volume (liters)

## Notes

(1) The concentration of this reagent can be checked by standard methods such as arsenious oxide titration method (A.O.A.C., 6.139, 1945).

(2) The pH of the solution should be less than 2.0. The yellow or brown color of $I_2$ should not be present.

(3) A total of four extractions should suffice, for adequate removal of iodine into the organic layer. The color of the iodine-carbon tetrachloride layer may be used to judge the degree of extraction.

(4) A 10 ml or 20 ml lusteroid centrifuge tube or suitable glass or plastic vial may be used.

## Reference

1. Glendenin, L. E., and Metcalf, R. P., Paper 278 in "Radiochemical Studies: The Fission Products," Coryell, C. D. and Sugarman, N., (eds), p. 1625, McGraw-Hill, New York, 1951.

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11-16-59

# VI. APPENDIX

## REAGENT PREPARATION

### A. CARRIERS*

Barium carrier, 70 mg/ml. Dissolve 132 grams of $Ba(NO_3)_2$ in 800 ml of water and dilute to a volume of 1 liter with distilled water.

Barium carrier, 20 mg/ml. Dissolve 38 grams of $Ba(NO_3)_2$ in 800 ml of water and dilute to a volume of 1 liter with distilled water.

Barium carrier, 7 mg/ml. Dilute 10 ml of barium carrier (70 mg/ml) to 100 ml with distilled water.

Cerium carrier, 10 mg/ml. Dissolve 29 grams of $Ce(NO_3)_3 \cdot 6\ H_2O$ in 800 ml of water and dilute to a volume of 1 liter with distilled water.

Cesium carrier, 20 mg/ml. Dissolve 29 grams of $CsNO_3$ in 800 ml of water and dilute to a volume of 1 liter with distilled water.

Cobalt carrier, 20 mg/ml. Dissolve 84.7 grams $Co(C_2H_3O_2)_2 \cdot 4H_2O$ in water and dilute to 1 liter with distilled water.

Iodine carrier, 13 mg/ml. Dissolve 15 grams of NaI in 900 ml of water and dilute to 1 liter with distilled water.

Iron carrier, 20 mg/ml. Dissolve 9 grams $FeCl_3 \cdot 6H_2O$ in water and dilute to 1 liter with distilled water.

Lead carrier, 100 mg/ml. Dissolve 165.6 grams of $Pb(NO_3)_2$ in water and dilute to 1 liter with distilled water.

Lead carrier, 50 mg/ml. Dissolve 82.8 grams of $Pb(NO_3)_2$ in water and dilute to 1 liter with distilled water.

Mixed carrier solution, Sr, 22 mg; Cs, 3.25 mg; Fe, 1.25 mg; Zr, 1.25 mg; Zn, 1.25 mg; Ce, 1.25 mg, and Co, 1.25 mg/ml. Dissolve 105.8 grams $Sr(NO_3)_2$; 9.75 grams $CsNO_3$; 12.2 grams $FeCl_3 \cdot 6H_2O$; 8.9 grams $ZrOCl_2 \cdot 8H_2O$; 11.39 grams $Zn(NO_3)_2 \cdot 6H_2O$; 7.74 grams $Ce(NO_3)_3 \cdot 6H_2O$; and 10.56 grams $Co(C_2H_3O_2)_2 \cdot 4H_2O$ in 700 ml of distilled water. Add 5 ml of 12 N HCl and dilute to 1 liter with distilled water.

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* Where the exact concentration is necessary for determination of chemical yield standardization is necessary.

Niobium carrier, 20 mg/ml. Dissolve 26 grams $K_8Nb_6O_{19}\cdot 16H_2O$ in 800 ml 1 N $HNO_3$. Filter, then dilute to 1 liter with 1 N $HNO_3$.

Niobium carrier, 10 mg/ml. Dissolve 13 grams $K_8Nb_6O_{19}\cdot 16H_2O$ in 800 ml 1 N $HNO_3$. Filter, then dilute to 1 liter with 1 N $HNO_3$.

Rare earth carrier, mixed. Dissolve 12.8 grams $Ce(NO_3)_3\cdot 6H_2O$; 14 grams $ZrOCl_2.8H_2O$, and 25 grams $FeCl_3\cdot 6H_2O$ in 600 ml of water containing 5 ml of 12 N HCl and dilute to 1 liter with distilled water.

Strontium carrier, 88 mg/ml. Dissolve 211 grams of $Sr(NO_3)_2$ in 700 ml of water and dilute to 1 liter with distilled water.

Strontium carrier, 20 mg/ml. Dissolve 48 grams of $Sr(NO_3)_2$ in 700 ml of water and dilute to 1 liter with distilled water.

Strontium, barium, and cesium carrier, mixed. Dissolve 21.2 grams $Sr(NO_3)_2$; 26.1 grams $Ba(NO_3)_2$, and 19.5 grams of $CsNO_3$ in distilled water and dilute to 1 liter with distilled water.

Zinc carrier, 20 mg/ml. Dissolve 91 grams $Zn(NO_3)_2\cdot 6H_2O$ in water and dilute to 1 liter with distilled water.

Zirconium carrier, 10 mg/ml. Dissolve 30 grams $ZrO(NO_3)_2\cdot 2H_2O$ in 1 N $HNO_3$. Filter, dilute to 1 liter with 1 N $HNO_3$.

B. OTHER REAGENTS

Acetic acid, 18 N. Concentrated reagent, glacial 99.5 per cent.

Acetic acid, 1.5 N. Add 86 ml of glacial acetic acid to 800 ml of water and dilute to a volume of 1 liter with distilled water.

Ammonium acetate, 3 N. Dissolve 231 grams of $NH_4C_2H_3O_2$ in 600 ml of water and dilute to a volume of 1 liter with distilled water.

Ammonium acetate buffer, pH 5.0. Mix 100 ml of 1.5 N acetic acid and 200 ml of 3 N ammonium acetate.

Ammonium acetate wash solution. Dilute 30 ml of ammonium acetate buffer with 200 ml of distilled water.

Ammonium hydroxide, 15 N. Concentrated reagent, aqua ammonia, specific gravity 0.90.

Ammonium hydroxide, 6 N. Add 400 ml of 15 N $NH_4OH$ to 400 ml of water and dilute to 1 liter with distilled water.

Ammonium molybdate, 0.5 N. Dissolve 100 grams of $(NH_4)_6Mo_7O_{24}\cdot 4H_2O$ in 800 ml of water and dilute to 1 liter with distilled water.

Ammonium oxalate, saturated. Dissolve 60 grams of $(NH_4)_2C_2O_4\cdot H_2O$ in 450 ml of hot distilled water.

Ammonium oxalate wash solution, 0.5 per cent. Dissolve 5 grams of $(NH_4)_2C_2O_4.H_2O$ in 1 liter of distilled water.

Anion exchange resin, strong basic form ($Cl^-$) x 8, 50 - 100 mesh. Wash slowly with 3 column volumes of ethanol, followed by distilled water, 4 or 5 column volumes of 6 N $NH_4OH$, distilled water, 4 or 5 column volumes of 6 N HCl, and finally by distilled water.

Cation exchange resin, strong acid form ($H^+$) x 8, 50 - 100 mesh. Wash slowly with 3 column volumes of ethanol, followed by distilled water, 4 or 5 column volumes of 6 N $NH_4OH$, distilled water, 4 or 5 column volumes of 6 N HCl, and finally by distilled water.

Citric acid, 3 N. Dissolve 192 grams of $H_3C_6H_5O_7$ in 600 ml of water, add 5 ml of 0.1 per cent phenol to prevent biological growth and dilute to a volume of 1 liter with distilled water.

Cupferron, 6 per cent. Dissolve 6 grams of cupferron in 100 ml of $H_2O$ and filter.

Ethylenediaminetetraacetic acid, (EDTA) M/4. Dissolve 73 grams EDTA in distilled water and dilute to 1 liter with distilled water.

Hydrochloric acid, 12 N. This is HCl specific gravity 1.19.

Hydrochloric acid, 6 N. Add 500 ml of 12 N HCl to 400 ml of water and dilute to 1 liter with distilled water.

Hydrochloric acid, 1 N. Add 84 ml of 12 N HCl to 800 ml of water and dilute to 1 liter with distilled water.

Hydrofluoric acid, 1 N. Add 36 ml of 28 N HF to 800 ml of water and dilute to 1 liter with distilled water.

Hydrofluoric acid, hydrochloric acid, mixture. Add 11 ml of 28 N HF to 666 ml of 12 N HCl.

Iodic acid, 0.35 N. Dissolve 10.5 grams $HIO_3$ in water and dilute to 1 liter with distilled water.

Methyl orange, 0.1 per cent. Dissolve 0.5 grams of methyl orange in 500 ml of distilled water.

Nitric acid, 16 N. This is concentrated reagent, specific gravity 1.42.

Nitric acid, 14 N. Add cautiously 900 ml of 16 N $HNO_3$ to 100 ml of distilled water.

Nitric acid, 8 N. Add cautiously 500 ml of 16 N $HNO_3$ to 400 ml of water and dilute to 1 liter with distilled water.

Nitric acid, 4 N. Add cautiously 250 ml of 16 N $HNO_3$ to 700 ml of water and dilute to 1 liter with distilled water.

Nitric acid, 1 N. Add cautiously 64 ml of 16 N $HNO_3$ to 900 ml of water and dilute to 1 liter with distilled water.

Nitric acid, 0.1 N. Add 6.4 ml 16 N $HNO_3$ to 900 ml of water and dilute to 1 liter with distilled water.

Potassium chromate, 1.0 N. Dissolve 98 grams of $K_2CrO_4$ in 800 ml of water and dilute to 1 liter with distilled water.

8-quinolinol solution, 7.5 per cent. Dissolve 75 grams of 8-quinolinol in 800 ml of methanol and dilute to 1 liter with methanol.

Sodium bisulfite, 1 N. Dissolve 52 grams $NaHSO_3$ in 800 ml of water and dilute to 1 liter with distilled water. Sodium meta-bisulfite ($Na_2S_2O_5$) may be substituted (48 grams per liter).

Sodium carbonate, 3 N. Dissolve 160 grams of $Na_2CO_3$ in 600 ml of water and dilute to 1 liter with distilled water.

Sodium chromate, 3 N. Dissolve 513 grams of $Na_2CrO_4 \cdot 10H_2O$ in 400 ml of water and dilute to 1 liter with distilled water.

Sodium chromate, 1 N. Dissolve 171 grams of $Na_2CrO_4 \cdot 10H_2O$ in 600 ml of water and dilute to 1 liter with distilled water.

Sodium cobaltinitrite reagent. Dissolve 230 grams of $NaNO_2$ in 500 ml $H_2O$, add 165 ml 6 N acetic acid, and 30 grams $Co(NO_3)_2 \cdot 6H_2O$, let stand overnight, filter, and dilute to 1 liter. This is 0.3 N $Na_3Co(NO_2)_6$, 3 N $NaNO_2$, 1 N acetic acid.

Sodium hydroxide, 19 N. Dissolve 760 grams of NaOH in 700 ml of water and dilute to 1 liter with distilled water.

Sodium hypochlorite, 5.25 per cent. Commercial household laundry bleach.

Sodium nitrate, 1 N. Dissolve 85 grams of $NaNO_3$ in 800 ml of water and dilute to 1 liter with distilled water.

Sodium nitrite, 1 N. Dissolve 69 grams $NaNO_2$ in 800 ml of water and dilute to 1 liter with distilled water.

Sulfuric acid, 18 N. Add cautiously with stirring 500 ml of 36 N $H_2SO_4$ to 100 ml of water and dilute to 1 liter with distilled water.

Tributyl phosphate, reagent grade, equilibrated with 14 N $HNO_3$. Mix thoroughly for 10 minutes equal parts of TBP and 14 N $HNO_3$. Discard the aqueous layer.

# CALCULATION OF $Sr^{90}$ ACTIVITY FROM $Y^{90}$ COUNTING

This is governed by the equation for ingrowth of $Y^{90}$ in $Sr^{90}$

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^{\,o} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^{\,o} e^{-\lambda_2 t} \qquad 1)^{*}$$

where the subscript 2 refers to the daughter ($Y^{90}$) and 1 to the parent ($Sr^{90}$), the superscript o refers to zero time, N = Number of atoms, $\lambda$ = decay constant, and t = time, and by the decay equation for Y-90:

$$N_2 = N_2^{\,o} e^{-\lambda_2 t} \qquad 2)$$

By making the following assumptions:

(a) $\lambda_1 << \lambda_2$,

(b) Over a time t of a few days or weeks, $\lambda_1 t \doteq 0$,

(c) $N_2^{\,o} = 0$,

equation (1) can be simplified into:

$$N_2 = \frac{\lambda_1}{\lambda_2} N_1^{\,o} (1 - e^{-\lambda_2 t}) \qquad 3)$$

Multiplying both equations (3) and (2) throughout by $\lambda_2$ gives the activity $A_2$, since $A = \lambda N$.

$$A_2 = A_1^{\,o} (1 - e^{-\lambda_2 t}) \quad \text{(ingrowth)} \qquad 4)$$

$$A_2 = A_2^{\,o} e^{-\lambda_2 t} \quad \text{(decay)} \qquad 5)$$

With this as introduction, the calculation can be illustrated by an example.

---

* Friedlander, G., and Kennedy, J. W., Nuclear and Radiochemistry, p. 130, John Wiley and Sons, Inc., New York, (1955).

Data:

| | | |
|---|---|---|
| Time of removal of rare earths | 3 PM | January 3 |
| Time of final TBP extraction | 10 AM | January 6 |
| Time of counting Y-90 | 1 PM | January 6 |

Ingrowth Time = 67 hours

and Decay Time = 3 hours

Measured Counting Rate of Y-90, R = 5.0 cpm (net)

Efficiency for Y-90 Counting $\eta = 0.75$ (75%)

A: Calculation of Y-90 activity, when counted ($A_2$):

$$A_2 = R_2 / \eta_2 = \frac{5.0}{0.75} = 6.7 \text{ dpm}$$

B: Calculation of Y-90 activity, when extracted ($A_2^o$):

$$A_2 = A_2^o e^{-\lambda_2 t}, \text{ or}$$

$$A_2^o = A_2 e^{\lambda_2 t} = (6.7) e^{(.693)(3)/(64)} = (6.7)(1.033) = 6.9 \text{ dpm}$$

C: Calculation of Sr-90 activity:

$$A_2 = A_1^o (1 - e^{-\lambda_2 t})$$

$$6.9 = A_1^o \left[ 1 - e^{-\frac{(.693)(67)}{64}} \right] = A_1^o (1 - .484)$$

$$A_1^o = \frac{6.9}{0.516} = 13.4 \text{ dpm}$$

$$= \frac{13.4}{2.22} \approx 6.0 \ \mu\mu c$$

# SAMPLE FORMS, COUNTING SHEETS, ETC.

SHEET # _____

OBSERVER _____

SOURCE _____

PLATEAU SHEET

RADIOLOGICAL HEALTH PROGRAM

SEC. - USPHS

DATE _____

INST. # _____

| TIME | SCALE FACTOR | REGISTERS | LIGHTS | C/MIN. | VOLTAGE | % | REMARKS |
|---|---|---|---|---|---|---|---|
| | | | | | | | |
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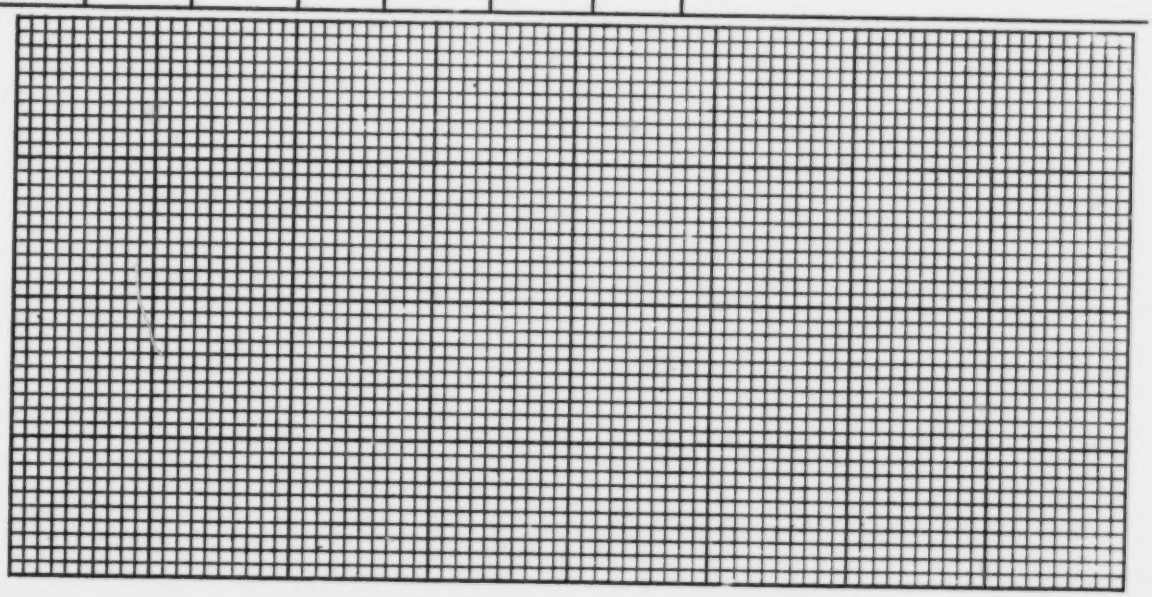
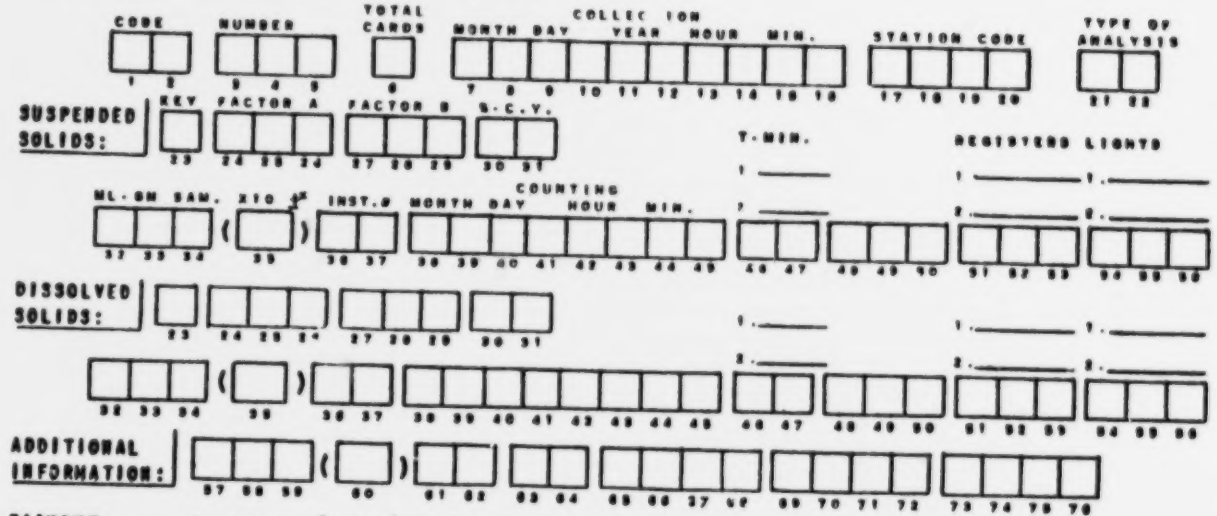


SEC-299
(12-58)

# RECORD FOR RADIOACTIVITY ANALYSES - RHRA




RAINOUT.....L. COLL. (X10$^{-}$) BEGAN..ENDED..AREA(X.XXX)..mg/L____SS...mg/L____DS...
BWQ NETWORK.....mg/L____SS...mg/L____DS...
AIR--Cs4.....
CISTERN.....W-ADDED---(X10$^{-}$) WAT-D % COLL.....mg/L____SS...mg/L____DS...
BIOLOGICAL....Gm. COLL...(X10$^{-}$).....%DRY WT(XX.XX)%ASH WT(XX.XX)
RAD. CH-NON...L. COLL...(X10$^{-}$).....mg/L____SS....g/L____DS....
Sr-90.....AMT.COLL..(X10$^{-}$).....

## SAMPLE ROUTING

DATE REC'D _____ BY _____ DATE COLL. _____ HOUR _____ SAMPLE TYPE _____ CODE NUMBER _____ STATION _____

| ROUTED TO: | ANALYSES NEEDED: | ON THE DS | SS | TS | DK | SPECIAL INSTRUCTIONS | COMPLETED AS OF | BY | PERSONAL REFERENCE | REPORTED TO: | d a a | REMARKS |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | GROSS-α | | | | | | | | | | | |
| 2 | GROSS-β | | | | | | | | | | | |
| 3 | γ-SCAN | | | | | | | | | | | |
| CHEMISTRY: | | | | | | | | | | | | |
| 4 | Sr-90 | | | | | | | | | | | |
| 5 | Sr-89,90 | | | | | | | | | | | |
| 6 | Cs-137 | | | | | | | | | | | |
| 7 | Ba-140 | | | | | | | | | | | |
| 8 | TRITIUM | | | | | | | | | | | |
| 9 | T.R.E. | | | | | | | | | | | |
| 10 | | | | | | | | | | | | |
| 11 | | | | | | | | | | | | |
| 12 | | | | | | | | | | | | |
| 13 | | | | | | | | | | | | |
| 14 | | | | | | | | | | | | |
| PHYSICS | | | | | | | | | | | | |
| 15 | I-131 | | | | | | | | | | | |
| 16 | | | | | | | | | | | | |
| 17 | | | | | | | | | | | | |
| 18 | | | | | | | | | | | | |
| 19 | | | | | | | | | | | | |

SEC - 75
(3 - 58)

# SAMPLE COUNTING DATA RECORD
## RADIOLOGICAL HEALTH PROGRAM
## SEC-USPHS

PROJECT: _____ OPERATING VOLTAGE: _____ DATE _____

BOOK NO.: _____ INSTRUMENT NO. _____

SHEET NO. _____

BACKGROUND: _____ CPM

| SAMPLE | HOUR | TIME MIN. | SCALE FACTOR | REG. READ | LIGHT COUNT | GROSS CPM | NET CPM | 95% E | OBSER. | REMARKS |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | | |
| | | | | | | | | | | |
| | | | | | | | | | | |

SEC 236
(11-58)

# RECORD OF
# LOW BACKGROUND (ANTI-COINCIDENCE) BETA COUNTER

OBSER: _____ DATE: _____

PROJECT: _____ INSTRUMENT NO. _____

BOOK NO.: _____ SHEET NO. _____

UMBRELLA "C" VOLTAGE: _____

## SAMPLE COUNTING DATA

COUNTER "A" VOLTAGE: _____ COUNTER "B" VOLTAGE: _____ "C"

| SAMPLE | GROSS | | NET CPM | 95% E | HOUR | TIME MIN. | SAMPLE | GROSS | | NET CPM | 95% E | GROSS COUNT |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | COUNT | CPM | | | | | | COUNT | CPM | | | |
| | | | | | | | | | | | | |
| | | | | | | | | | | | | |
| | | | | | | | | |

SEC 309
(8-59)

SAMPLE NOS. _____ TO _____

# LABORATORY DATA OF STRONTIUM $^{90}$ DETERMINATION

SR SEPN DATE _____

| (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) | (10) | (11) | (12) | (13) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| SAMPLE NO. DATE COLLECTED | SR SEPN DATE / HOUR | SR EXTN DATE / HOUR | CTG DATE / HOUR | GROSS CPM TIME(M) | BKGD CPM TIME(M) | NET CPM DPM* | INGRO TIME DAYS FACTOR | DECAY TIME HOURS FACTOR | SR YIELD | VOL(L) WT(G) | μμC/l μμC/g | 95% E |
| | | | | | | | | | | | | |
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CALCULATION

$$\frac{\text{DPM}}{(\text{COL 8})(\text{COL 9})(\text{COL 10})(\text{COL 11})\,2.22} = \text{COL 12}$$

* (LBBC), $\frac{\text{cpm}}{0.396}$

(IPC), $\frac{\text{cpm}}{0.75}$

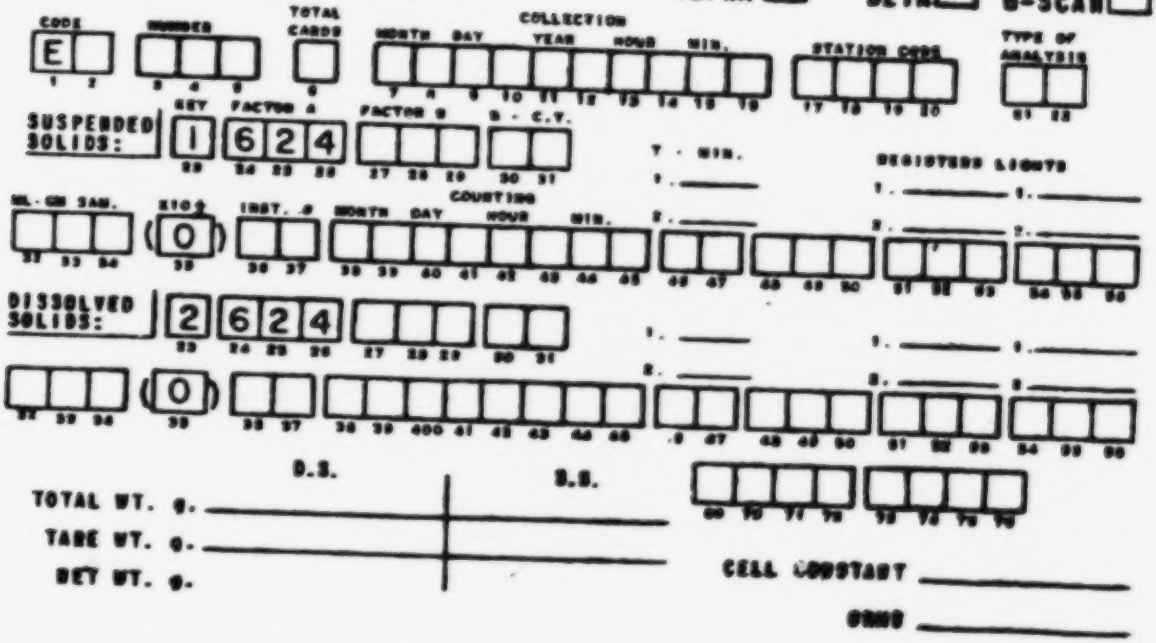
| SAMPLE NO. | DRY WT. 110° C GRAMS | ASH WT. GRAMS | CaO WT GRAMS | SPECIAL ANALYSES WT. GRAMS |
|---|---|---|---|---|
| | GROSS | GROSS | GROSS | GROSS |
| | TARE | TARE | TARE | TARE |
| | NET | NET | NET | NET |
| | GROSS | GROSS | GROSS | GROSS |
| | TARE | TARE | TARE | TARE |
| | NET | NET | NET | NET |
| | GROSS | GROSS | GROSS | GROSS |
| | TARE | TARE | TARE | TARE |
| | NET | NET | NET | NET |
| | GROSS | GROSS | GROSS | GROSS |
| | TARE | TARE | TARE | TARE |
| | NET | NET | NET | NET |

SEC 320
(9-69)

# RECORD FOR BASIC WATER QUALITY RADIOACTIVITY ANALYSES - RHRA

STATION _____
DATE REC'D _____

ALPHA ☐ BETA ☐ G-SCAN ☐



SEC-65
(6-58)

RECORD CARD FOR NATIONAL AIR SAMPLING NETWORK DATA

Station _____ Anal. Cat. _____ Filter No. _____

Time(min) _____ Sampling Rate(CuM/min) _____ Air Volume(CuM) _____

| Particulate: | | Organic | | β Radioactivity: | |
|---|---|---|---|---|---|
| Filter+Sample | _____ | Tube+Sample | _____ | Date | CPM |
| Filter | _____ | Tube | _____ | _____ | _____ |
| Sample | _____ | Sample | _____ | _____ | _____ |
| | | Total Wt. | _____ | _____ | _____ |

| Station Identification | Col. No. | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | Reflectance: _____ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Code No. | | | | | | | | | | | |

| | Month | | Day | | WD WE | Year | | Filter No. | | | | | Particulate µg/CuM | | | | Organic µg/CuM | | | | β Radact. µµc/CuM | | | | | Det. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Col. No. | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 | 29 | 30 | 31 | 32 | 33 | 34 | 35 | D.R. |
| Card 1 | | | | | | | | | | | | | | | | | | | • | | | | | • | | $T_{\frac{1}{2}}$ |
| Card 2 | | | | | | | | | | | | | | | | | x | x | x | x | 27 | 28 | 29 | 30 | 31 | |

| | Starting Time | | | Hour Off. | |
|---|---|---|---|---|---|
| | Month | Day | Hour | | |
| Card 3 | 31 | 32 | 33 | 34 | 35 | 36 | 37 | 38 |

Filter No._____ Cu M_____ Sp. F._____

Card No. 1

| | D. Rdg. | | μg/Cu M | | | | | | Card Col. |
|---|---|---|---|---|---|---|---|---|---|
| Ba | | | x | x | | | | | 36-39 |
| Be | | | x | x | x | | | | 40-42 |
| Sulfate | | | | | | | x | x | 43-46 |
| Nitrate | | | | | | | x | x | 47-50 |
| pH | | | x | | | | x | x | 51-53 |
| Reflect. | | | x | | | o | x | x | 54-56 |
| Fluoride | | | x | x | | | | x | 57-59 |
| Chloride | | | x | | | | x | x | 60-62 |
| As | | | x | x | | | | x | 63-65 |
| Ca | | | x | | | | x | x | 66-68 |
| Mg | | | x | | | | x | x | 69-71 |
| Sr | | | x | x | | | | x | 72-74 |
| Na | | | x | | | | x | x | 75-77 |
| K | | | x | | | | x | x | 78-80 |

Card No. 2

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Mn | | | x | x | | | | x | 32-34 |
| Pb | | | x | | | | x | x | 35-37 |
| Sn | | | x | x | | | | x | 38-40 |
| Fe | | | x | | | | x | x | 41-43 |
| Cu | | | x | | | | | x | 44-47 |
| Ti | | | x | x | | | | x | 48-50 |
| V | | | x | x | | | | | 51-54 |
| Zn | | | x | | | | | x | 55-58 |
| Cr | | | x | x | x | | | | 59-61 |
| Ni | | | x | x | | | | | 62-65 |
| Mo | | | x | x | | | | x | 66-68 |
| Co | | | x | x | x | | | | 69-71 |
| Bi | | | x | x | x | | | | 72-74 |
| Cd | | | x | x | x | | | | 75-77 |
| Sb | | | x | x | x | | | | 78-80 |

Card No. 3

| Date | Time | Visual Range | Restrictions | | | | Sky Condition | |
|---|---|---|---|---|---|---|---|---|
| 1 | | | | | | | | 39-45 |
| 2 | | | | | | | | 46-52 |
| 3 | | | | | | | | 53-59 |
| 1 | | | | | | | | 60-66 |
| 2 | | | | | | | | 67-73 |
| 3 | | | | | | | | 74-80 |